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A Lagrangian Analysis of Turbulent Diffusion

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This is an analysis of diffusion of a scalar field by molecular transport and isotropic turbulence. Existing results are surveyed, and some new results are advanced. The discussion is supported with oceanographic and atmospheric observations of dispersion and diffusion. The existing results were originally obtained using a variety of mathematical techniques. However, all results are derived here using an approximate solution of the Lagrangian form of the advection-diffusion equation. The approximation is equivalent to neglecting the spatial dependence of the transformation factors in the Lagrangian representation of the molecular flux divergence. Examinations of the diffusive subranges show the approximation to justified: infinitesimal line stretching is either controlled by relatively large scale shears (viscous-diffusive subrange at large Prandtl number) or else is negligible during the diffusion process (inertia-diffusive subrange at small Prandtl number). Estimation of scalar mean fields, total variances, and wave number spectra requires, in general, joint statistics of infinitesimal line stretching and either single particle displacement or particle pair separation. Normality is assumed for displacement statistics; separation statistics are determined from the Richardson-Kraichnan equation. A simple derivation of that equation is presented here. Joint stretching-separation statistics are modeled by a uniform shear flow, with time-dependent amplitudes described by the Wiener process (white noise). With the possible exception of this random process, the only mathematics required here is elementary calculus, so details have been kept to a minimum. In the diffusion problems considered here, the turbulence is isotropic. However, both the approximate solution of the advection-diffusion equation and the equations for joint displacements are equally valid for inhomogeneous turbulence.

CONTENTS

1. INTRODUCTION

The widespread use of drifters in meteorology [e.g., *Julian et al.*, 1977; *Er-El and Peskin*, 1981] and oceanography [e.g., *Freeland et al.*, 1975; *Colin de Verdiere*, 1983; *Royer and Emery*, 1984; *Davis*, 1985; *Rossby et al.*, 1985, and references therein; *Garrett et al.*, 1985] requires an examination of recent analytical theories of turbulent diffusion. In particular, it is important to be able to use a knowledge of the turbulent dispersion of marked fluid particles in order to estimate concentrations of dissolved, passive scalar substances which are subject to the combined effects of turbulent dispersion and molecular diffusion ("turbulent diffusion"). Here molecular diffusion may also be interpreted as the mixing effects of small-scale turbulence or waves, as parameterized by *Young et al.* [1982], for example.

Turbulent diffusion has attracted an enormous amount of attention, and it is fortunate that there are a number of texts, proceedings, and reviews on the subject written for the benefit of meteorologists and oceanographers [*Csanady*, 1973; *Fischer et al.*, 1979; *Harris*, 1979; *Okubo*, 1980; *Pasquill and Smith*, 1983; *Chatwin and Allen*, 1985; *Hunt*, 1985]. These all contain careful discussions of the fundamentals, such as the random-walk model of particle displacement. They also contain detailed analyses of difficult but important problems of diffusion in anisotropic and inhomogeneous turbulence, such as planetary boundary layers and coastal waters. However, the material in these texts appears greatly removed from that which appears in the later sections of the encyclopedic treatise by *Monin and Yaglom* [1975, sections 23 and 24] and even further from the highly sophisticated theoretical analyses which have appeared in the recent literature. The latter include renormalized series expansions [*Kraichnan*, 1966, 1977; *Phythian and Curtis*, 1978], stochastic differential equations [*Durbin*, 1980], and Feynman path integrals [*Drummond*, 1982].

The purpose of this article is to survey the results of the theoretical analyses, using a unified but simple analytical approach which, it is hoped, has the same spirit and level of complexity of the texts mentioned above. The results include the various subrange forms for the wave number spectra of scalar variance in isotropic turbulence. There is substantial observational support for almost all these forms. The results also include total scalar variances, due to several source con-

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figurations. Some of these results lack observational support as yet, while others are controversial to the extent that the analysis given here does not agree with that given elsewhere. Lastly, there is a discussion of the total effective diffusivity for the mean scalar field; it is argued that there is uncertainty even as to the existence of such a quantity.

An essential feature of the approach here is the use of an approximate solution of the advection-diffusion equation in Lagrangian coordinates, thereby emphasizing the hydrodynamic aspect of the physics and the relationship with fluid particle kinematics. This emphasis is at odds with the mathematical perspective in which the equation is of parabolic type, rather than hyperbolic type, irrespective of the smallness of the nondimensional diffusion coefficient or Peclet number.

The approximate solution is obtained by neglecting the spatial dependence of the transformation factors in the Lagrangian representation of the diffusion operator. This is equivalent to assuming that the flow has uniform shear. However, the uniform shear assumption is not made, in general, when calculating other Lagrangian quantities such as particle displacements and separation. The approximate solution permits the formulation of explicit expressions for mean scalar concentrations, total scalar variance, and scalar wave number spectra. Evaluation of these expressions requires, in general, a knowledge of the statistics of the stretching of infinitesimal elements, jointly with the statistics of either the finite separation between particle pairs or else the displacement of their centroid.

In most of the processes here it suffices to know just the marginal statistics of displacement or separation. There is substantial theoretical support [*Cocke*, 1972] and observational support [e.g., *Davis*, 1985] for the hypothesis that displacement statistics are asymptotically normal for large time, with mean and variance in accordance with the classical theory of *Taylor* [1921] in the special case of stationary homogeneous turbulence. There is also a strong attraction toward displacement probability distribution functions (pdfs) which possess the Markov or group property, at least asymptotically for large time. This property guarantees weak forms of Corrsin's hypotheses, relating Lagrangian statistics at different labeling times. It is also necessary that the marginal statistics, for the displacement of one of two particles, be independent of the second particle. With these three a priori requirements of asymptotic normality, marginality, and the Markov property, approximate evolution equations for displacement and separation pdfs are developed here. The approximations are in the form of discards of certain triple correlations. There are several ways of making such discards, but apparently only one meets the a priori requirements.

For isotropic turbulence the separation pdf so derived is the Richardson-Kraichnan equation [*Richardson*, 1926; *Kraichnan*, 1966; *Lundgren*, 1981]. The relative diffusivities appearing in the equation are deduced from their spectral representations using dimensional and scaling arguments, in various subranges, for small and large times. Dimensional arguments alone suffice for large times but not for the small time appropriate in inertia-diffusive subranges. On the other hand, the estimation of diffusivities, or equivalently, Lagrangian velocity correlations, is all that is necessary in some processes: the separation pdf itself is not required.

In other processes, such as the viscous-diffusive subrange, joint statistics of stretching and separation are required. These statistics are modeled using uniform shear flow models. The shear amplitudes have various time dependences: constants [*Batchelor*, 1959] and white noise or Wiener processes [*Kraichnan*, 1974]. The latter process is the most sophisticated mathematical concept invoked here. Comprehensive descriptions for physicists may be found in the work by *van Kampen* [1981]. Mathematical details have been avoided in this article, especially as only elementary calculus is involved. The most detail is in section 2, on Lagrangian formulation. The processes examined in sections 3–6 would appear to be in order of decreasing complexity, from variance spectra to mean fields. In fact, the results progress from well established to highly speculative. Oceanic and atmospheric observations are used to support theoretical developments. Data range from temperature microstructure in fjords to the dispersion of stratospheric balloons. Results are briefly summaried in section 8, with emphasis on the novel aspects. There is a brief allusion to recent work on anisotropic and wave induced diffusion.

2. LAGRANGIAN FORMULATION

2.1. *Formulation*

Let C denote the concentration of a passive scalar substance. It is required to find C at some position $\mathbf{x}$ in N-dimensional space and at some time t. The concentration $C(\mathbf{x}, t)$ is determined by the linear advection-diffusion equation, a source distribution, and initial values.

It will be convenient to use $\mathbf{X}$ and s as Eulerian dummy variables. In terms of these variables the evolution equation for C is

$$\frac{\partial C}{\partial s} + \mathbf{u} \cdot \nabla C = \kappa \nabla^2 C + S \tag{1}$$

where the gradient operator is

$$\nabla = \partial/\partial X_i \qquad i = 1, 2, \cdots, N$$

the advecting velocity $\mathbf{u} = \mathbf{u}(\mathbf{X}, s)$ is solenoidal,

$$\nabla \cdot \mathbf{u} = 0$$

the source $S = S(\mathbf{X}, s)$ varies in space and time, and κ is a constant molecular diffusivity. For simplicity alone it will be assumed that initially C vanishes:

$$C(\mathbf{X}, 0) = 0 \tag{2}$$

For each realization of the turbulent velocity field $\mathbf{u}$, and for each space-time point $(\mathbf{x}, t)$, there is a particle path $\mathbf{A}(\mathbf{x}, t|s)$. That is, $(\mathbf{X}, s)$ lies on the path if and only if

$$\mathbf{X} = \mathbf{A}(\mathbf{x}, t \,|\, s)$$

The function $\mathbf{A}$ is determined by

$$D\mathbf{A}/Ds = \mathbf{u}(\mathbf{A}, s) \tag{3}$$

subject to

$$\mathbf{A}(\mathbf{x}, t \,|\, t) = \mathbf{x}$$

So the particle passes through $\mathbf{X} = \mathbf{x}$ at $s = t$. The derivative (D/Ds) denotes differentiation with respect to s, with $(\mathbf{x}, t)$ fixed. Note the inverse functional relationship

$$\mathbf{x} = \mathbf{A}(\mathbf{X}, s \,|\, t)$$

In terms of the Lagrangian coordinates ($\mathbf{x}$, t) and s, and for a realization of $\mathbf{u}$, C becomes

$$C(\mathbf{X}, s) = C[\mathbf{A}(\mathbf{x}, t \mid s), s] \equiv C(\mathbf{x}, t \mid s)$$

in *Kraichnan*'s [1965] notation.

The Eulerian equation (equation (1)) may be expressed in terms of the Lagrangian coordinates. The left-hand side is just the ("total") derivative of $C(\mathbf{x}, t \mid s)$ with respect to s:

$$\left[\frac{\partial C}{\partial s}(\mathbf{X}, s) + u_j(\mathbf{X}, s)\frac{\partial C}{\partial X_j}(\mathbf{X}, s)\right]_{\mathbf{X}=\mathbf{A}(\mathbf{x}, t|s)} = \frac{D}{Ds} C[\mathbf{A}(\mathbf{x}, t \mid s), s]$$

by virtue of (3) and the chain rule for derivatives. In order to express the right-hand side in Lagrangian form, use the chain rule in the form

$$\frac{\partial}{\partial X_i} C(\mathbf{X}, s) = \frac{\partial}{\partial X_i} C[\mathbf{A}(\mathbf{X}, s \mid t), t \mid s]$$

$$= \frac{\partial}{\partial x_j} C(\mathbf{x}, t \mid s) \frac{\partial A_j}{\partial X_i}(\mathbf{X}, s \mid t)$$

This may be repeated to obtain a Lagrangian expression for second derivatives of $C(\mathbf{X}, s)$ with respect to $\mathbf{X}$. Hence the Lagrangian form of (1) is

$$\frac{DC}{Ds} = \kappa \nabla A_i \cdot \nabla A_j \frac{\partial^2 C}{\partial x_i \partial x_j} + \kappa \nabla^2 A_i \frac{\partial C}{\partial x_i} + S \tag{4}$$

where the summation convention has been adopted, and $S = S(\mathbf{x}, t \mid s)$. The detailed forms of the transformation factors are

$$(\nabla A_i)_k \equiv \frac{\partial A_i}{\partial X_k} \equiv \left[\left(\frac{\partial}{\partial X_k}\right) A_i(\mathbf{X}, s \mid t)\right]_{\mathbf{X}=\mathbf{A}(\mathbf{x}, t|s)}$$

$$\nabla^2 A_i \equiv \frac{\partial^2 A_i}{\partial X_k \partial X_k} \equiv \left[\left(\frac{\partial^2}{\partial X_k \partial X_k}\right) A_i(\mathbf{X}, s \mid t)\right]_{\mathbf{X}=\mathbf{A}(\mathbf{x}, t|s)}$$

Like (1), (4) is of advection-diffusion type. The vector $\nabla^2 \mathbf{A}$ acts like an advecting velocity, but it is not solenoidal. The vectors ∇A_i alter the rate of diffusion: when $s = t$ they are the unit vectors $\hat{\mathbf{e}}_i$, but on average they increase as $|t - s|$ increases, since, as it will be seen, they evolve in the same way as infinitesimal line elements. This is a Lagrangian expression of the familiar statement that turbulence enhances the effective rate of molecular dissipation of C, by transferring the variance of C to small scales. In the Lagrangian formulation (equation (4)) the simplicity of the convective derivative (D/Ds) is achieved at the expense of complicating the diffusion operator. Equation (4) is no more readily integrated than is (1).

2.2. *Solution*

In order to proceed, the x dependence of the transformation factors in (4) will be neglected. This assumption will be justified in subsequent sections using one of the following arguments:

1. Diffusion of the scalar C is negligible.
2. Scalar diffusion is much faster than infinitesimal line stretching: equilibrium between the external scalar source and the diffusion sink is attained before the transformation factors have altered significantly from their spatially uniform initial values.
3. Line stretching is principally due to velocity shears with scales much larger than those at which scalar diffusion is significant; thus stretching rates are approximately uniform in space.
4. Only upper bounds for the scalar variance are required, and adequate bounds are obtained without having to admit line stretching.

The advantage of the above assumption is that (4) is then readily integrated subject to the initial condition (equation (2)) [*Okubo et al.*, 1983], yielding

$$C(\mathbf{x}, t) = C(\mathbf{x}, t \mid t) = \int_0^t ds \int d\mathbf{y}\, G(\mathbf{x} - \mathbf{y}, t, s) S(\mathbf{y}, t \mid s) \tag{5}$$

The fundamental solution G is most clearly expressed by its Fourier transform g:

$$g(\mathbf{k}, t - s) = \exp\left\{-\kappa \int_s^t [\mathbf{k} \cdot \mathbf{Q} \cdot \mathbf{k} - i\mathbf{k} \cdot \mathbf{L}]\, da\right\} \tag{6a}$$

where

$$Q_{ij} = \nabla A_i \cdot \nabla A_j \tag{6b}$$

$$L_i = \nabla^2 A_i \tag{6c}$$

$$\nabla A_i = \nabla A_i(\ \ , 0 \mid a) \qquad \nabla^2 A_i = \nabla^2 A_i(\ \ , 0 \mid a) \tag{6d}$$

$$g(\mathbf{k}, t, s) = \int d\mathbf{x} e^{i\mathbf{k}\cdot\mathbf{x}} G(\mathbf{x}, t, s) \tag{6e}$$

The suppressed argument in (, $0 \mid a$) indicates that the $\mathbf{x}$ dependence has been ignored. In fact, the arguments (, $t \mid t - a$) should be used, but statistical stationarity and time reversibility will be assumed in subsequent sections. The solution (5) is represented graphically in Figure 1, which shows the path $[A(\mathbf{y}, t \mid s), s]$ of a parcel of fluid and the diffusion cloud $G(\mathbf{x} - \mathbf{y}, t, s)$. It is emphasized that particle paths will not necessarily be determined by assuming that the velocity field has uniform shear.

Now that an explicit, albeit model-approximate, representation has been obtained for the scalar concentration $C(\mathbf{x}, t)$, its statistics may be calculated directly.

2.3. *Variance Spectrum*

Assume that the velocity field $\mathbf{u}$ and source distribution S are independent, isotropic, and stationary random fields, both with vanishing means (although alternative assumptions about S will be made in some later sections). Ensemble averages over one or both fields will be denoted $\langle\ \rangle_u$, $\langle\ \rangle_S$, or $\langle\ \rangle_{u,S}$. When there is no ambiguity, the subscripts will be dropped. For example,

$$\langle \mathbf{u} \rangle = \mathbf{0} \qquad \langle S \rangle = 0 \tag{7}$$

The representation of C provided by (5) may be used to calculate the spatial covariance of C, at spatial lag $\mathbf{r}$ and at absolute time t:

$$\langle C(\mathbf{x} + \mathbf{D}, t) C(\mathbf{x}, t) \rangle_{u,S} = \int_0^t ds_1 \int_0^t ds_2 \int d\mathbf{y}_1 \int d\mathbf{y}_2 \cdot \langle G(\mathbf{x} + \mathbf{D} - \mathbf{y}_1, t, s_1) G(\mathbf{x} - \mathbf{y}_2, t, s_2) \cdot \langle S(\mathbf{y}_1, t \mid s_1) S(\mathbf{y}_2, t \mid s_2) \rangle_S \rangle_u \tag{8}$$

In particular, V, the total variance of C, is just the covariance at zero lag:

$$V \equiv \langle C(\mathbf{x}, t)^2 \rangle_{u,S} \tag{9}$$

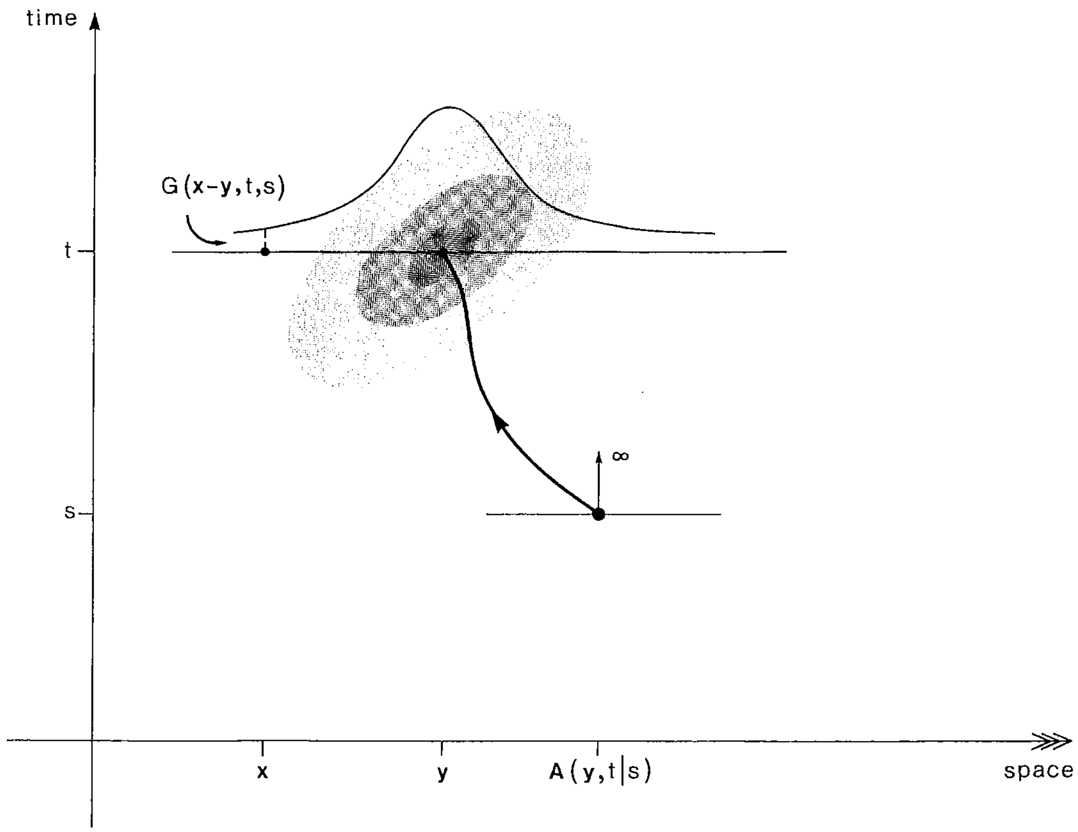

Fig. 1. Graphical representation of the solution (5). The scalar injected by the source S at $\mathbf{A}(\mathbf{y}, t\,|\,s)$, at time s, has diffused into an ellipsoidal cloud about $\mathbf{y}$ at time t. The cloud concentration at $\mathbf{x}$ at time t is proportional to $G(\mathbf{x} - \mathbf{y}, t, s)$. The vertical arrow through $[\mathbf{A}(\mathbf{y}, t\,|\,s), s]$ indicates that $G(\mathbf{x} - \mathbf{y}, s, s) = \delta(\mathbf{x}, \mathbf{y})$. In this graphical representation the diffusing cloud has been rotated 90° out of the space manifold to which it properly belongs.

As a consequence of the isotropy of $\mathbf{u}$ and S, the covariance of C is a function only of $r = |\mathbf{r}|$ and t, while V is a function only of t.

The one-dimensional wave number spectrum of C will be derived as the Fourier transform of the covariance of C:

$$F(k, t) = \int da\,(\mathbf{k}) \int d\mathbf{D}\; e^{i\mathbf{k}\cdot\mathbf{D}} \langle C(\mathbf{x} + \mathbf{D}, t) C(\mathbf{x}, t)\rangle_{u,S}$$

where $da\,(\mathbf{k})$ is an area element on the surface of a sphere of radius k. Since the covariance depends only upon r, the Fourier transform depends only upon k, and the integrals reduce to

$$F(k, t) = a(k) \int_0^\infty dD\; a(D)\mathscr{B}(kD)\langle C(\mathbf{x} + \mathbf{D}, t)C(\mathbf{x}, t)\rangle_{u,S} \tag{10}$$

where

$$a(k) = 2\pi k \qquad N = 2$$
$$a(k) = 4\pi k^2 \qquad N = 3 \tag{11}$$

$$\mathscr{B}(kD) = J_0(kD) \qquad N = 2$$
$$\mathscr{B}(kD) = \frac{\sin\,(kD)}{kD} \qquad N = 3 \tag{12}$$

The total variance V may be calculated as

$$V = (2\pi)^{-N} \int_0^\infty F(k, t)\; dk \tag{13}$$

The variance spectrum F is calculated by substituting the covariance representation (equation (8)) into the transform (equation (10)). The resulting formula includes the Lagrangian source covariance which may be expressed in Eulerian coordinates:

$$\begin{aligned}\langle S(\mathbf{y}_1, t\,|\,s_1)S(\mathbf{y}_2, t\,|\,s_2)\rangle_S &= \langle S[\mathbf{A}(\mathbf{y}_1, t\,|\,s_1), s_1]S[\mathbf{A}(\mathbf{y}_2, t\,|\,s_2), s_2]\rangle_S \\ &= V_S[|\mathbf{A}(\mathbf{y}_1, t\,|\,s_1) - \mathbf{A}(\mathbf{y}_2, t\,|\,s_2)|, s_1 - s_2]\end{aligned} \tag{14}$$

where $V_S(R, w)$ is the Eulerian source covariance at spatial lag $\mathbf{R}$ and time lag w:

$$V_S(R, w) \equiv \langle S(\mathbf{X} + \mathbf{R}, s + w)S(\mathbf{X}, s)\rangle \tag{15}$$

V_S depends on R and w only, since isotropy and stationarity have been assumed for S. A very simple model for V_S will be adopted:

$$V_S(R, w) = \chi\delta(w)\mathscr{B}(lR) \tag{16}$$

where δ is the Dirac delta function, $\mathscr{B}$ is defined in (12), and χ is a constant with the same dimensions as S^2t and C^2t^{-1}. That is, the source has a "white noise" time dependence [*Van Kampen*, 1981]. Since

$$a(k) \int d\mathbf{R}\; e^{i\mathbf{k}\cdot\mathbf{R}}\mathscr{B}(Rl) = a(k) \int_0^\infty dR\; a(R)\mathscr{B}(kR)\mathscr{B}(lR) = (2\pi)^N\delta(k - l) \tag{17}$$

the spatial structure of V_S corresponds to a source of scalar variance only at wave numbers $\mathbf{k}$ with magnitude $k = l$. It is the behavior of the scalar variance spectrum $F(k, t)$ for values of $k \gg l$ which is of interest, and this is independent of the details of the low wave number source. The model (equation (14)) simplifies some of the calculations.

In order to complete the evaluation of F, it remains to take the velocity field average $\langle\ \rangle_u$ in (8) and (10). The random variables which actually appear are the transformation or stretching factors $\mathbf{Q}$ and $\mathbf{L}$: see (6) and the single time particle separation $\mathbf{R} = \mathbf{A}(\mathbf{y}_1, t\,|\,s) - \mathbf{A}(\mathbf{y}_2, t\,|\,s)$. The two distinct times s_1 and s_2 need no longer be considered, since S is delta-correlated in time: see (14). Thus $\langle\ \rangle_u$ may be calculated by multiplying by the joint probability distribution function of the random variables $\mathbf{Q}$, $\mathbf{L}$, and $\mathbf{R}$ at time s (given respective values of unit matrix $\mathbf{I}$, $\mathbf{0}$, and $\mathbf{r} = \mathbf{y}_1 - \mathbf{y}_2$ at time t) and then integrating over $\mathbf{Q}$, $\mathbf{L}$, and $\mathbf{R}$. A little rearrangement using (6) yields

$$F(k, t) = \chi a(k) \int_0^t ds \int d\mathbf{r} \int d\mathbf{Q} \int d\mathbf{L} \int d\mathbf{R} \cdot e^{i\mathbf{k}\cdot\mathbf{r}} \mathscr{B}(lR) g^2(\mathbf{k}, t, s) P(\mathbf{Q}, \mathbf{L}, \mathbf{R}, s\,|\,\mathbf{I}, \mathbf{O}, \mathbf{r}, t) \tag{18}$$

Hence the determination of F is reduced to estimating the joint pdf of separation and stretching appearing in (18). This will be carried out in section 4 for each of the various subranges of F: inertia convective, viscous convective, inertia diffusive, and viscous diffusive.

Estimates are easier to obtain in the first three subranges, where g is approximately independent of the stretching variables $\mathbf{Q}$ and $\mathbf{L}$. Then the integration over those variables in (16) is trivial, leading to

$$F(k, t) = \chi a(k) \int_0^t ds \int d\mathbf{r} \int d\mathbf{R}\ e^{i\mathbf{k}\cdot\mathbf{r}} \mathscr{B}(lR) P(\mathbf{R}, s\,|\,\mathbf{r}, t) \tag{19}$$

where P is now the marginal pdf for the vector separation $\mathbf{R}$ at time s. Since $\mathbf{R}$ has the deterministic or statistically sharp value $\mathbf{r}$ at time t, it must be the case that $P(\mathbf{R}, t\,|\,\mathbf{r}, t) = \delta(\mathbf{R} - \mathbf{r}) = \delta(R_1 - r_1) \cdots \delta(R_N - r_N)$ where $\mathbf{R} = (R_1, \cdots, R_N)$ and $\mathbf{r} = (r_1, \cdots, r_N)$. In particular, $\int d\mathbf{R} P(\mathbf{R}, t\,|\,\mathbf{r}, t) = 1$, but this must also hold for all values of $s \leq t$.

It may be noted that P depends upon the vector $\mathbf{R}$ even though the turbulence is isotropic, since $\mathbf{r}$ has a direction (and vice versa). However, *Lundgren* [1981] pointed out that the spherically averaged pdf

$$P(R, s\,|\,r, t) = a(R)^{-1} \int da\,(\mathbf{R}) P(\mathbf{R}, s\,|\,\mathbf{r}, t) \tag{20}$$

must depend only upon r. The corresponding initial condition is

$$P(R, t\,|\,r, t) = a(r)^{-1}\delta(r - R) \tag{21}$$

and the normalization is

$$\int_0^\infty dR\ a(R) P(R, s\,|\,r, t) \equiv 1 \tag{22}$$

In terms of this P, the representation (equation (19)) for the variance spectrum F becomes

$$F(k, t) = \chi a(k) \int_0^t ds \int_0^\infty dr \int_0^\infty dR\ a(r) a(R) \cdot \mathscr{B}(kr) \mathscr{B}(lR) P(R, s\,|\,r, t) \tag{23}$$

The symbol P has now been used to denote three different pdfs. The displayed arguments indicate which one is involved. This is simpler, and more informative, than introducing a separate symbol for each pdf.

In order to use (21) and (23) to calculate $F(k, t)$, an evolution equation for P is required. Such an equation will be obtained in the next section. Before proceeding, however, it is instructive to examine (23). It is easily seen that

$$\frac{\partial F}{\partial t}(k, t) = (2\pi)^N \chi \delta(k - l) + \mathscr{I}(k, t) \tag{24}$$

where $\mathscr{I}$ is the integral in (23) with P replaced by $(\partial P/\partial t)$. The first term on the right-hand side of (24) represents the source of scalar variance at wave number l. The term $\mathscr{I}$ represents turbulent transfer of variance to wave number k from other wave numbers. It is also easily shown that $\int_0^\infty \mathscr{I}(k, t)\, dk \equiv 0$, implying conservation of variance in the absence of a source. Clearly, development of an evolution equation for the separation pdf $P(R, s\,|\,r, t)$ is equivalent to developing a model for the spectral transfer rate $\mathscr{I}(k, t)$.

3. SEPARATION PDF

As was mentioned at the end of the preceding section, much can be inferred from the marginal statistics for the separation of particle pairs. Specifically, the (scalar) separation of particle pairs. Specifically, the (scalar) separation pdf $P(R, s\,|\,r, t)$ is required, for $s \leq t$. The pdf is known at $s = t$ (see (21)), so an evolution equation is required. Such an equation will be derived in this section. The derivation will proceed in stages. The main result is in subsection 3.4.

3.1. *Displacement of a Single Particle*

A particle passing through the point $\mathbf{X}$ at time s also passes through $\mathbf{x} = \mathbf{A}(\mathbf{X}, s\,|\,t)$ at time t, where $\mathbf{A}$ is defined by (3) and is determined by a given realization of the turbulent velocity field $\mathbf{u}$. For this realization the "micro" pdf of $\mathbf{x}$ at time t is

$$p(\mathbf{x}, t\,|\,\mathbf{X}, s) = \delta[\mathbf{x} - \mathbf{A}(\mathbf{X}, s\,|\,t)] \tag{25}$$

Note that the order of the arguments of p is the reverse of that required in section 2. The reason is that p will be found to satisfy, approximately, a diffusion equation for which it is natural to take the larger of t and s as the time variable. The required order of arguments will be obtained subsequently.

For incompressible flow, p satisfies the Liouville equation

$$\frac{\partial p}{\partial t} + u_i \frac{\partial p}{\partial x_i} = 0 \tag{26}$$

where $u_i = u_i(\mathbf{x}, t)$. Hence the "macro" pdf

$$P(\mathbf{x}, t\,|\,\mathbf{X}, s) \equiv \langle \delta[\mathbf{x} - \mathbf{A}(\mathbf{X}, s\,|\,t)] \rangle \tag{27}$$

satisfies

$$\frac{\partial P}{\partial t} + \left\langle u_i \frac{\partial p}{\partial x_i} \right\rangle = 0 \tag{28}$$

The initial condition is

$$P(\mathbf{x}, s\,|\,\mathbf{X}, s) = \delta(\mathbf{x} - \mathbf{X}) \tag{29}$$

The problem is the estimation of the flux term in (28). To this end, equations are derived for P and $p' = p - P$:

$$\frac{\partial P}{\partial t} + \langle u_i \rangle \frac{\partial P}{\partial x_i} = -\frac{\partial}{\partial x_i} \langle u_i' p' \rangle \tag{30}$$

$$\frac{\partial p'}{\partial t} + u_i \frac{\partial p'}{\partial x_i} = T_1(\mathbf{x}, t) + T_2(\mathbf{x}, t) \tag{31}$$

where $u_i'(\mathbf{x}, t) = u_i(\mathbf{x}, t) - \langle u_i(\mathbf{x}, t) \rangle$, and

$$T_1 = \left\langle u_j' \frac{\partial p'}{\partial x_j} \right\rangle \qquad T_2 = -u_j' \frac{\partial P}{\partial x_j} \tag{32}$$

The solution of (31) is

$$p' = \sum_{k=1}^{2} \int_s^t T_k[\mathbf{A}(\mathbf{x}, t \mid v), v] \, dv \tag{33}$$

which may be used to express the random flux $\langle u_i' p' \rangle$ in (30). So far, this analysis is exact. Two approximations will now be made:

1. In the contribution to the random flux arising from T_1, the arguments $[\mathbf{A}(\mathbf{x}, t \mid v), v]$ are replaced by $(\mathbf{x}, t)$. Consequently, this contribution is proportional to $\langle u_i'(\mathbf{x}, t) \rangle$, which vanishes. In other words, we neglect the correlation between $u_i(\mathbf{x}, t)$ and $T_1[\mathbf{A}(\mathbf{x}, t \mid v), v]$.

2. In the contribution to the random flux arising from T_2, the arguments $[\mathbf{A}(\mathbf{x}, t \mid v), v]$ of $\partial P/\partial x_i$ are replaced by $(\mathbf{x}, t)$, while $u_i'(\mathbf{x}, t \mid v)$ is replaced by $u_i(\mathbf{x}, t \mid v)'$. These approximations amount to neglecting triple Lagrangian correlations and yield

$$\langle u_i' p_i' \rangle = -\int_s^t \langle u_i'(\mathbf{x}, t) u_j(\mathbf{x}, t \mid v)' \rangle \, dv \frac{\partial P}{\partial x_j}(\mathbf{x}, t \mid \mathbf{X}, s) \tag{34}$$

and so

$$\frac{\partial P}{\partial t} + \langle u_i(\mathbf{x}, t) \rangle \frac{\partial P}{\partial x_i} = \frac{\partial}{\partial x_i}\left[D_{ij}(\mathbf{x}, \mathbf{x}, t \mid s) \frac{\partial P}{\partial x_j} \right] \tag{35}$$

where the Lagrangian diffusivity D_{ij} is the integral in (34).

This diffusion equation has an attractive property. For stationary turbulence, $\langle u_i \rangle$ is independent of time while $D_{ij}(\mathbf{x}, \mathbf{x}, t \mid s) = D_{ij}(\mathbf{x}, \mathbf{x}, t - s) \to D_{ij}(\mathbf{x}, \mathbf{x}, \infty)$ as $t - s \to \infty$. That is, (35) has the asymptotic form

$$\partial P/\partial t \sim \mathscr{D} P \tag{36}$$

where $\mathscr{D}$ is a differential operator with respect to $\mathbf{x}$, independent of time t. The asymptotic solution for P is

$$P \sim e^{(t-s)\mathscr{D}} \delta(\mathbf{x} - \mathbf{X}) \tag{37}$$

The exponentiated operator should be interpreted as a power series. It readily follows that asymptotically, P satisfies the Markov property [*van Kampen*, 1981]:

$$P(\mathbf{x}, t \mid \mathbf{X}, s) \sim \int P(\mathbf{x}, t \mid \mathbf{Y}, v) P(\mathbf{Y}, v \mid \mathbf{X}, s) \, d\mathbf{Y} \tag{38}$$

as $v - s$ and $t - v \to \infty$. Equation (38) yields an estimate for the Lagrangian mean velocity:

$$\langle \mathbf{u}(\mathbf{X}, s \mid t) \rangle = \int \mathbf{x} \frac{\partial P}{\partial t}(\mathbf{x}, t \mid \mathbf{X}, s) \, d\mathbf{x} \tag{39}$$

Substituting (38) yields

$$\langle \mathbf{u}(\mathbf{X}, s \mid t) \rangle \sim \int \langle \mathbf{u}(\mathbf{Y}, v \mid t) \rangle P(\mathbf{Y}, v \mid \mathbf{X}, s) \, d\mathbf{Y} \tag{40}$$

which relates Lagrangian mean velocities. There are analogous expressions relating Lagrangian covariances. These are examples of weak forms of Corrsin's hypotheses [*Corrsin*, 1959]. In the strong form, (40) is assumed to hold for $v = t$ rather than as $t - v \to \infty$. Then $\langle \mathbf{u}(\mathbf{Y}, v \mid t) \rangle = \langle \mathbf{u}(\mathbf{Y}, t \mid t) \rangle = \langle \mathbf{u}(\mathbf{Y}, t) \rangle$, the Eulerian mean velocity field. The strong form of (40) can be derived directly from (35) and (39), provided $\partial D_{ij}/\partial x_j$ is negligible in comparison with $\langle u_i(\mathbf{x}, t) \rangle$; that is, the turbulence is only weakly inhomogeneous. On the other hand, approximations 1 and 2 which led to (35) could only be expected to hold for such turbulence. Finally, consider incompressible, stationary homogeneous turbulence. Then D_{ij} is independent of $\mathbf{x}$, and P is multivariate normal, with mean $(t - s)\langle u_i \rangle$ and covariance $\int_s^t D_{ij}(v) \, dv$, in accordance with the classical theory of *Taylor* [1921].

It is appropriate to discuss alternatives to approximation 2 used in the derivation of (35).

2*. Suppose instead in the contribution to the random flux term arising from T_2, the arguments $[\mathbf{A}(\mathbf{x}, t \mid v), v]$ are replaced by $(\mathbf{x}, t)$ everywhere. The resulting Eulerian diffusivity is

$$D_{ij}^* = (t - s) \langle u_i'(\mathbf{x}, t) u_j'(\mathbf{x}, t) \rangle$$

and the resulting pdf cannot possess the Markov property as $t - s \to \infty$.

2**. Alternatively, suppose in the contribution to the random flux term arising from T_2, the arguments $[\mathbf{A}(\mathbf{x}, t \mid v), v]$ of $\partial P/\partial x_i$ are replaced with $(\mathbf{x}, t)$ while those of u_i are replaced with $(\mathbf{x}, v)$. The resulting Eulerian diffusivity is

$$D_{ij}^{**} = \int_s^t \langle u_i'(\mathbf{x}, t) u_j'(\mathbf{x}, v) \rangle \, dv$$

The resulting pdf will possess the Markov property asymptotically for stationary turbulence and will be normal for stationary homogeneous turbulence, but the mean and covariance will not be in accordance with the classical Taylor theory.

Higher-order approximations to (35) may be found in the waves by *Kraichnan* [1977] and *Jiang* [1985]. The slightly novel derivation of (35) given here must be about as simple as can be.

3.2. *Joint Displacement of a Pair of Particles*

Consider a pair of particles which pass through $\mathbf{X}$ and $\mathbf{Y}$ at time s. The macro pdf for passage through $\mathbf{x}$ and $\mathbf{y}$, respectively, at time t is

$$P(\mathbf{x}, \mathbf{y}, t \mid \mathbf{X}, \mathbf{Y}, s) = \langle \delta[\mathbf{x} - \mathbf{A}(\mathbf{X}, s \mid t)] \delta[\mathbf{y} - \mathbf{A}(\mathbf{Y}, s \mid t)] \rangle \tag{41}$$

Proceeding as in the above subsection yields

$$\begin{aligned} \frac{\partial P}{\partial t} &+ \langle u_i(\mathbf{x}, t) \rangle \frac{\partial P}{\partial x_i} + \langle u_i(\mathbf{y}, t) \rangle \frac{\partial P}{\partial y_i} \\ &= \frac{\partial}{\partial x_i} \left\{ D_{ij}(\mathbf{x}, \mathbf{x}, t \mid s) \frac{\partial P}{\partial x_j} + D_{ij}(\mathbf{x}, \mathbf{y}, t \mid s) \frac{\partial P}{\partial y_j} \right\} \\ &+ \frac{\partial}{\partial y_i} \left\{ D_{ij}(\mathbf{y}, \mathbf{x}, t \mid s) \frac{\partial P}{\partial x_j} + D_{ij}(\mathbf{y}, \mathbf{y}, t \mid s) \frac{\partial P}{\partial y_j} \right\} \end{aligned} \tag{42}$$

where

$$D_{ij}(\mathbf{x}, \mathbf{y}, t \mid s) = \int_s^t \langle u_i'(\mathbf{x}, t) u_j(\mathbf{y}, t \mid v)' \rangle \, dv \tag{43}$$

Equation (42) was deduced by *Lundgren* [1981] in a superficially different manner. However, (42) is unacceptable, because the marginal equation obtained by integrating (42) over $\mathbf{y}$ is not the same as (35), the equation for the marginal pdf $P(\mathbf{x}, t \mid \mathbf{X}, s)$. For example, if (42) is used to obtain the Lagrangian drift of one particle, the result depends upon the presence of the second particle. It is also readily shown that (42) predicts a nonzero mean (vector) separation rate for particle pairs in homogeneous turbulence, which is clearly false.

The shortcomings of (42) may be remedied. For if we return the second prime in (43) to its original position, it follows that

$$\frac{\partial}{\partial y_j}\langle u_i'(\mathbf{x}, t)u_j'(\mathbf{y}, t \mid v)\rangle$$

$$= \left\langle u_i'(\mathbf{x}, t)\left(\frac{\partial u_j'}{\partial x_k}\right)(\mathbf{y}, t \mid v)\left(\frac{\partial A_k}{\partial y_j}\right)(\mathbf{y}, t \mid v)\right\rangle$$

$$\cong \left\langle u_i'(\mathbf{x}, t)\left(\frac{\partial u_j'}{\partial x_k}\right)(\mathbf{y}, t \mid v)\right\rangle\left\langle\left(\frac{\partial A_k}{\partial y_j}\right)(\mathbf{y}, t \mid v)\right\rangle \quad (44)$$

provided we neglect two-point triple correlations, which is consistent with the approximate derivation of (42). Moreover, if the turbulence is nearly homogeneous, then the right-hand side of (44) is approximately

$$\left\langle u_i'(\mathbf{x}, t)\left(\frac{\partial u_j'}{\partial x_k}\right)(\mathbf{y}, t \mid v)\right\rangle\delta_{kj}$$

$$= \left\langle u_i'(\mathbf{x}, t)\left(\frac{\partial u_j'}{\partial x_j}\right)(\mathbf{y}, t \mid v)\right\rangle = 0 \quad (45)$$

for incompressible flow. Consequently, the mixed terms in (42) may be replaced with more satisfactory forms:

$$D_{ij}(\mathbf{x}, \mathbf{y}, t \mid s)\frac{\partial P}{\partial y_j} \to \frac{\partial}{\partial y_j}\{D_{ij}(\mathbf{x}, \mathbf{y}, t \mid s)P\}$$

$$D_{ij}(\mathbf{y}, \mathbf{x}, t \mid s)\frac{\partial P}{\partial x_j} \to \frac{\partial}{\partial x_j}\{D_{ij}(\mathbf{y}, \mathbf{x}, t \mid s)P\} \quad (46)$$

Then the single-particle marginal equation is the same as (35). For weakly inhomogeneous turbulence, all of the derivatives in (42) may be moved to the left of the diffusivities. The resulting equation is then the same as that of *Kraichnan* [1965] except that in the latter, only the solenoidal parts of the Lagrangian velocities appear in the diffusivities. For example, $u_i'(\mathbf{x}, t \mid v)$ is replaced by $u_i'^S(\mathbf{x}, t \mid v)$ where

$$\frac{\partial}{\partial x_i}\{u_i'^S(\mathbf{x}, t \mid v)\} = 0$$

Such a replacement permits moving the derivatives in the desired manner.

Our approximation (equation (45)) is invalid for strongly inhomogeneous turbulence. Such a flow may be characterized by a mean strain rate Λ greatly in excess of the root-mean-square strain rate. Hence the displacement of a particle by the mean flow, through the eddy field, grows as exp $[\Lambda(t - s)]$. Therefore pairs of particles will be moving independently as soon as $t - s = O(\Lambda^{-1})$, almost irrespective of their initial separation, and so

$$P(\mathbf{x}, \mathbf{y}, t \mid \mathbf{X}, \mathbf{Y}, s) \sim P(\mathbf{x}, t \mid \mathbf{X}, s)P(\mathbf{y}, t \mid \mathbf{Y}, s) \quad (47)$$

which obviates the need for an evolution equation for the two-particle pdf.

3.3. *Separation of a Pair of Particles*

For isotropic turbulence the diffusivity tensors are solenoidal in both indices, and so (42) and its variants discussed above are identical. Let centroid and separation coordinates be defined by

$$\mathbf{c} = \tfrac{1}{2}(\mathbf{x} + \mathbf{y}) \qquad \mathbf{r} = \mathbf{x} - \mathbf{y}$$

$$\mathbf{C} = \tfrac{1}{2}(\mathbf{X} + \mathbf{Y}) \qquad \mathbf{R} = \mathbf{X} - \mathbf{Y}$$

Then the pair pdf becomes $P(\mathbf{r}, \mathbf{c}, t \mid \mathbf{R}, \mathbf{C}, s)$, and the marginal pdf for vector separation

$$P(\mathbf{r}, t \mid \mathbf{R}, s) = \int P(\mathbf{r}, \mathbf{c}, t \mid \mathbf{R}, \mathbf{C}, s)\, d\mathbf{c} \quad (48)$$

is independent of $\mathbf{C}$ by homogeneity. It is straightforward to derive, from (42), the following evolution equation for the marginal pdf:

$$\frac{\partial P}{\partial t} = \frac{\partial^2}{\partial r_i \partial r_j}(\eta_{ij}P) \quad (49)$$

where

$$\eta_{ij}(\mathbf{r}, t \mid s) = D_{ij}(\mathbf{x}, \mathbf{x}, t \mid s) - D_{ij}(\mathbf{x}, \mathbf{y}, t \mid s)$$

The initial condition for (49) is

$$P(\mathbf{r}, s \mid \mathbf{R}, s) = \delta(\mathbf{r} - \mathbf{R}) \quad (50)$$

Next, we may average the marginal pdf over the direction of $\mathbf{R}$ to obtain a pdf which, since the turbulence is isotropic, depends only on the magnitude $r = |\mathbf{r}|$:

$$P(r, t \mid R, s) = a(R)^{-1}\int P(\mathbf{r}, t \mid \mathbf{R}, s)\, da\,(\mathbf{R}) \quad (51)$$

which obeys

$$\frac{\partial P}{\partial t} = a(r)^{-1}\frac{\partial}{\partial r}\left[a(r)\eta\frac{\partial P}{\partial r}\right] \quad (52)$$

subject to

$$P(r, s \mid R, s) = a(R)^{-1}\delta(r - R) \quad (53)$$

where $\eta(r, t \mid s)$ is the longitudinal component of the incompressible isotropic tensor η_{ij} [*Batchelor*, 1960, section 3.4]. *Richardson* [1926] virtually guessed (52); he did not consider time dependence for η. *Kraichnan* [1965, 1966, Equation (3.6)] derived (52) using his Lagrangian history direct interaction approximation. *Lundgren* [1981] derived (52) by only assuming a velocity field delta correlated in time. This assumption is equivalent to making approximations (1) and (2) in subsection 3.1 above. By using the arguments of *Batchelor* [1960], the longitudinal diffusivity component η is expressible as

$$\eta(r, t \mid s) = a(r)^{-1}\int_0^r \zeta(\rho, t \mid s)\frac{d}{d\rho}a(\rho)\, d\rho \quad (54)$$

where the relative diffusivity ζ is defined by

$$\zeta(r, t \mid s) = \frac{D}{Dt}\langle r^2\rangle = 2\int_s^t dv\, \{\langle\mathbf{u}(\mathbf{x}, t)\cdot\mathbf{u}(\mathbf{x}, t \mid v)\rangle$$

$$- \langle\mathbf{u}(\mathbf{x}, t)\cdot\mathbf{u}(\mathbf{x} + \mathbf{r}, t \mid v)\rangle\} \quad (55)$$

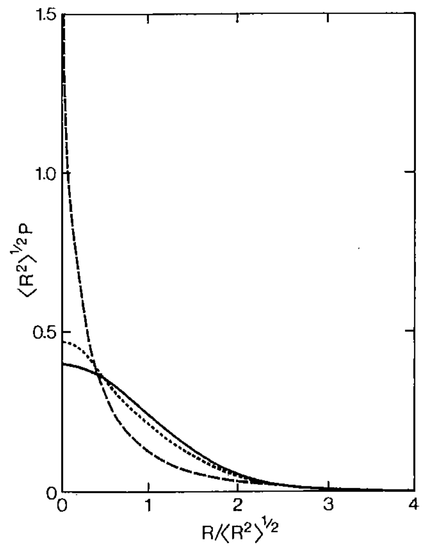

Fig. 2. Theoretical and observed separation pdfs: relative diffusivity independent of R [*Batchelor*, 1952] (solid line); relative diffusivity proportional to $R^{4/3}$ [*Richardson*, 1926] (dashed line); relative diffusivity inferred from observations of dye concentration in Lake Huron [after *Sullivan*, 1971] (dotted line).

Finally, note that (52) and (53) ensure the normalization

$$\int_0^{\infty} a(r)P(r, t \mid R, s)\, dr \equiv 1 \tag{56}$$

provided

$$a(r)\eta \frac{\partial P}{\partial r} \rightarrow 0 \qquad r \rightarrow 0 \quad r \rightarrow \infty$$

3.4. *Reversibility*

Our expression (equation (23)) for the scalar concentration spectrum $F(k, t)$ requires a knowledge of the scalar separation pdf $P(R, s \mid r, t)$ for values of the "running" time s less than the conditioning or initial time t. However, the diffusion equation (equation (52)) should only be integrated for running times greater than the conditioning time. *Lundgren* [1981] obtained an important result which resolves this problem. It is well known that for incompressible flow the Jacobian determinant of the transformation $\mathbf{X} \rightarrow \mathbf{x} = \mathbf{A}(\mathbf{X}, s \mid t)$ has unit magnitude. Hence

$$\delta[\mathbf{X} - \mathbf{A}(\mathbf{x}, t \mid s)] = \delta[\mathbf{x} - \mathbf{A}(\mathbf{X}, s \mid t)] \tag{57}$$

and so

$$P(\mathbf{X}, s \mid \mathbf{x}, t) = P(\mathbf{x}, t \mid \mathbf{X}, s) \tag{58}$$

Similarly,

$$P(\mathbf{X}, \mathbf{Y}, s \mid \mathbf{x}, \mathbf{y}, t) = P(\mathbf{x}, \mathbf{y}, t \mid \mathbf{X}, \mathbf{Y}, s) \tag{59}$$

and so

$$P(\mathbf{R}, \mathbf{C}, s \mid \mathbf{r}, \mathbf{c}, t) = P(\mathbf{r}, \mathbf{c}, t \mid \mathbf{R}, \mathbf{C}, s) \tag{60}$$

$$P(\mathbf{R}, s \mid \mathbf{r}, t) = P(\mathbf{r}, t \mid \mathbf{R}, s) \tag{61}$$

and finally,

$$P(R, s \mid r, t) = P(r, t \mid R, s) \tag{62}$$

It follows that for $s \leq t$, $P(R, s \mid r, t)$ satisfies

$$\frac{\partial P}{\partial s} = -a(R)^{-1} \frac{\partial}{\partial R}\left[a(R)\eta(R, s \mid t) \frac{\partial P}{\partial R}\right] \tag{63}$$

subject to

$$P(R, t \mid r, t) = a(r)^{-1}\delta(R - r) \tag{64}$$

Note that (63) is a "backward" diffusion equation. Once again, this result is a consequence of incompressibility alone. Neither stationarity nor homogeneity is necessary.

3.5. *Uniformity of the Approximate Theory*

Consider the approximate evolution equation (equation (35)) for the single particle displacement pdf. If the turbulence is inhomogeneous, then the Eulerian mean velocity $\langle u_i(\mathbf{x}, t)\rangle$ and the diffusivity $D_{ij}(\mathbf{x}, \mathbf{x}, t \mid s)$ are functions of $\mathbf{x}$, and in general, the pdf is not normal even as $(t - s) \rightarrow \infty$. This is in conflict with *Cocke* [1972], who proved a central limit theorem for integrals such as

$$\mathbf{A}(\mathbf{x}, t \mid s) = \mathbf{x} + \int_s^t \mathbf{u}(\mathbf{x}, t \mid v)\, dv \tag{65}$$

under general conditions which appear to include inhomogeneity and nonstationarity. The derivation of (35) began with the definition (equation (25)) of the micro pdf, which, for incompressible turbulence, satisfies a Liouville equation with Eulerian velocity $u_i(\mathbf{x}, t)$. However, the micro pdf also satisfies a Liouville equation with Lagrangian velocity $u_i(\mathbf{X}, s \mid t)$. The difference is superficial until an approximation is made for the random flux term in the equation for the averaged or macro pdf. The approximations which led to (35) would in the latter case lead to

$$\frac{\partial P}{\partial t} + \langle u_i(\mathbf{X}, s \mid t)\rangle \frac{\partial P}{\partial x_i} = K_{ij}(\mathbf{X}, \mathbf{X}, s \mid t) \frac{\partial^2 P}{\partial x_i \partial x_j} \tag{66}$$

where

$$K_{ij}(\mathbf{X}, \mathbf{X}, s \mid t) = \int_s^t \langle u_i(\mathbf{X}, s \mid t)' u_j(\mathbf{X}, s \mid v)'\rangle\, dv \tag{67}$$

The solution of (66) is exactly multivariate normal for all $(t - s)$, with mean and covariance in agreement with the classical Taylor theory, even for inhomogeneous nonstationary turbulence. There is an analogous equation for the pdf of joint displacements of particle pairs, also with exactly normal solutions. However, the utility of the Richardson-Kraichnan equation (equation (52)) lends some credence to (42), for homogeneous turbulence and $(t - s)$ not large. Thus neither of the two choices of velocity labeling in the Liouville equations leads to uniformly valid approximations. There must be a generalized coordinate $[Z(\mathbf{x}, t \mid \mathbf{X}, s), v(\mathbf{x}, t \mid \mathbf{X}, s)]$ which behaves like $(\mathbf{x}, t)$ for $(t - s)$ less than a velocity decorrelation time and like $[\mathbf{A}(\mathbf{x}, t \mid s), s]$ for $(t - s) \rightarrow \infty$.

3.6. *Stochastic Models*

There is a model for (49). That is, (49) is the forward Fokker-Planck equation [*van Kampen*, 1981] for the Ito stochastic differential equation

$$d\mathbf{r} = \xi^{1/2}\, d\boldsymbol{\omega}(t) \tag{68}$$

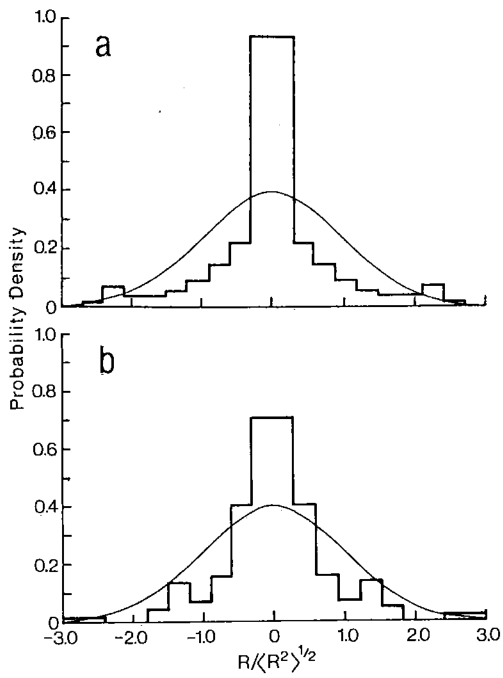

Fig. 3. Sample pdfs for (*a*) zonal and (*b*) meridional components of separations of high-altitude balloons, 5 days after release in the southern hemisphere subtropics. The normal pdfs are included for reference. The kurtoses are 7.54 and 7.02, respectively [after *Er-El and Peskin*, 1981].

where $\xi_{ij} = \eta_{ij} + \eta_{ji}$ and the components of $\boldsymbol{\omega}(t)$ are independent Wiener processes. The tensor $\boldsymbol{\xi}$ is symmetric so (68) is meaningful if $\boldsymbol{\xi}$ is nonnegative. In the Itō interpretation, $\boldsymbol{\xi} = \boldsymbol{\xi}(\mathbf{r}, t\,|\,s)$, and so the nonlinear equation (68) is explicit. There are analogous Itō models for (42) and its variants. Numerical integration of the models may be a useful technique for determining the pdfs, or at least some of their moments, in anisotropic or inhomogeneous turbulence.

3.7. *Observations*

There have been few attempts to observe the separation pdf $P(R, s\,|\,r, t)$. Dye measurements (Figure 2) in Lake Huron [*Sullivan*, 1971] did not support Richardson's solution of (63), which is based on the assumption that $\eta \propto R^{4/3}$ (see section 4.4). The measurements were more consistent with a normal distribution for $\mathbf{R}$, which may be derived from (49) by assuming η_{ij} is independent of $\mathbf{R}$. This would be the case if the two particles were moving independently, with normally distributed displacements, that is, for an elapsed time greatly exceeding the turbulent decorrelation time.

The pdfs for the zonal and meridional components of separation of high-altitude balloons were estimated by *Er-El and Peskin* [1981], on the basis of 178 observations 5 days after launch. Significantly nonnormal pdfs were found, with kurtoses of 7.54 and 7.02, respectively (see Figure 3). For normal distributions the kurtosis has the value 3.

Surface drifters deployed off the California coast by *Davis* [1985] were used to estimate separation pdfs (see Figure 4). Pairs with initial separations in the range 16 km $< r <$ 30 km had separations R after 4 days, closely consistent with a normal distribution for $\mathbf{R}$. Those with initial separations in the range 4 km $< r <$ 16 km were more likely after 4 days to have small separations R than in the case of normally distributed $\mathbf{R}$. *Davis* [1985] attributes this result to trapping in small-scale velocity convergence or else to exponentially growing separations in a large-scale shear as discussed in section 32. Davis also presents data purporting to show that η does not depend on R alone but rather on R and $t - s$. However, it should be noted that what is shown is a dependence upon $\langle R^2\rangle^{1/2}$ rather than the conditional or deterministic value R. This point is also discussed in section 4.4.

4. CONVECTIVE SUBRANGES OF THE SCALAR VARIANCE SPECTRUM

The particle pair separation statistics described in the previous section will be used in this section to construct the convective subranges of the variance spectrum, that is, the subranges in which κ is so small that scalar diffusion may be neglected. Conditions under which this approximation fails (sufficiently high wave number) will be given in the next section, which concerns diffusive subranges.

In order to define the scalar subranges, it is first necessary to describe the subranges of the kinetic energy spectrum.

4.1. *Kinetic Energy Subranges*

The wave number spectrum for isotropic, stationary turbulence is

$$E(k) = a(k) \int_0^\infty dD\; a(\mathbf{D})\mathscr{B}(kD)\langle \mathbf{u}(\mathbf{x} + \mathbf{D}, t) \cdot \mathbf{u}(\mathbf{x}, t)\rangle \qquad (69)$$

It is assumed that this equilibrium spectrum is maintained by a statistically stationary source, at or around some low wave number m; the average source strength must match the average energy dissipation rate ε. The latter is dominated by viscous dissipation at high wave numbers. By assumption there are no sources or sinks at intermediate wave numbers, so the energy spectrum in such an "inertial" subrange can only depend on ε and the wave number k. By dimensional analysis,

$$E(k) = K_\gamma \varepsilon^{2/3} k^{-5/3} \qquad (70)$$

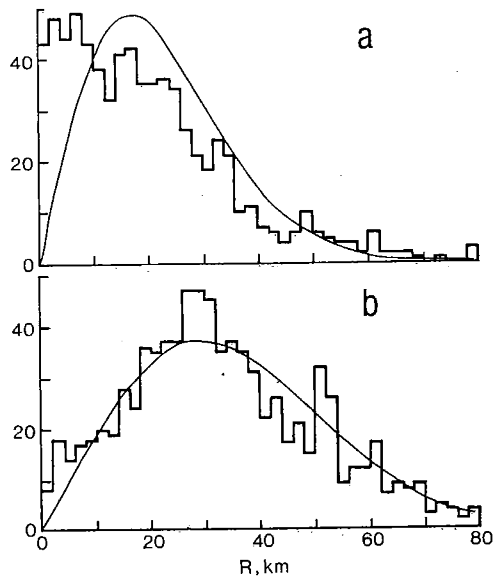

Fig. 4. Histograms of separations of ocean surface drifters, 4 days after release off the California coast: (*a*) initial separations 4 km $< r <$ 16 km and (*b*) initial separations 16 km $< r <$ 30 km. The histograms are based on bins 2 km wide. The smooth curves correspond to a normal distribution for $\mathbf{R}$. [after *Davis*, 1985].

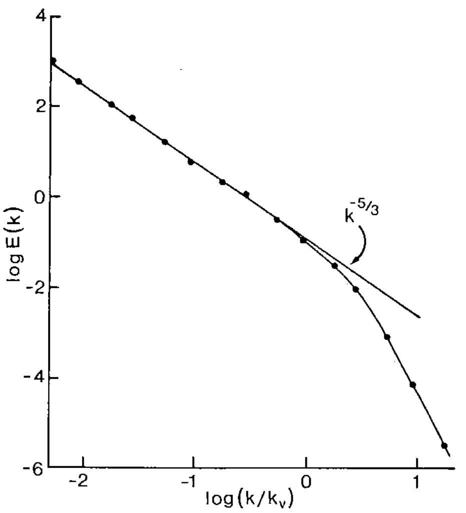

Fig. 5. Wave number spectrum of kinetic energy $E(k)$ in a British Columbia tidal channel. The Kolmogorov wave number $k_V = (\varepsilon/\nu^3)^{1/4}$ has the value 0.21 m, given $\varepsilon = 0.61 \times 10^{-4}$ m^2 s^{-1} and $\nu = 1.48 \times 10^{-6}$ m^2 s^{-1} (seawater at 8°C). The straight line has a slope of $-\frac{5}{3}$ [after *Grant et al.* 1962].

where K_V is the dimensionless Kolomogorov constant. For a comprehensive discussion, see *Monin and Yaglom* [1975]. The inertial time scale $\varepsilon^{-1/3}k^{-2/3}$ exceeds the viscous time scale $\nu^{-1}k^{-2}$ (where ν is the kinematic viscosity) if $k > k_V \equiv \varepsilon^{1/4}\nu^{-3/4}$. Thus (70) can hold only for $m \ll k \ll k_V$. If $k_V \ll k$, then E must depend upon ε and ν, and dimensional analysis will not suffice. Observations [*Grant et al.*, 1962] (Figure 5 here) indicate very rapid roll-off of $E(k)$ for $k \gg k_V$. At least one turbulence closure theory, the abridged Lagrangian history direct interaction approximation [*Kraichnan*, 1966], is in impressive agreement with the observations of the "dissipation range." It suffices, however, for our purposes to note only the very rapid decay for $k \gg k_V$, which will be modeled by a truncated form:

$$\begin{aligned} E(k) &= K_V\varepsilon^{-2/3}k^{-5/3} && k \le f k_V \\ E(k) &= 0 && k > f k_V \end{aligned} \tag{71}$$

where f is some fraction. The observations indicate $f \cong 0.1$.

As is indicated by the observations of *Grant et al.* [1962], the above description of isotropic turbulence is well substantiated in three space dimensions, but *Kraichnan* [1967] has proposed an alternative inertial subrange in two space dimensions. It is characterized by a statistically steady transfer of vorticity variance $\langle|\hat{\mathbf{e}}_3 \cdot \nabla \times \mathbf{u}|^2\rangle$ or enstrophy, from low to high wave numbers at a rate λ, which has the same dimensions as t^{-3}. By dimensional analysis the energy spectrum must be

$$E(k) = K_r\lambda^{2/3}k^{-3} \tag{72}$$

where K_r is a dimensionless constant, which it seems appropriate to name after Kraichnan. There is evidence of (72) in large-scale atmospheric circulation [*Boer and Shepherd*, 1983] (see Figure 6). As might be expected, that data does not survive the stringent tests for isotropy passed by smaller-scale data supporting (70) [*Gargett et al.*, 1984; *Gargett*, 1985]. The enstrophy inertial subrange should extend to wave numbers beyond which the flow cannot be described as two dimensional. *Young et al.* [1982] suggest that the upper limit may be $k \cong 10^{-3}$ m^{-1}.

4.2. *Convective Subranges*

In convective subranges the scalar variance spectrum $F(k, t)$ may be calculated using (23), which requires knowledge of the separation pdf $P(R, s\,|\,r, t)$. The latter will be determined using the backward Richardson-Kraichnan equation (equation (63)), subject to the initial or, more correctly, "final" condition (equation (64)). The longitudinal diffusivity $\eta(R, s\,|\,t)$ is given by (54) and (55), with the obvious change in notation; thus it is determined by the structure of the velocity field. It is possible to relate η to the energy spectrum and hence the energy subranges, since (54) and (55) may be expressed in the form [*Kraichnan*, 1966]

$$\eta(R, s\,|\,t) = 4\int_s^t dw \int_0^\infty dk\; E(k)\mathscr{F}(kR)\mathscr{L}(k, w - s) \tag{73}$$

where

$$\mathscr{F}(\theta) = a(\theta)^{-1}\int_0^\theta d\phi\; a(\phi)[1 - \mathscr{B}(\phi)] \tag{74}$$

is a high-pass filter arising from the geometry of isotropic turbulence,

$$\begin{aligned} \mathscr{F}(\theta) &\propto \theta^2 && \theta \to 0 \\ \mathscr{F}(\theta) &\to N^{-1} && \theta \to \infty \end{aligned} \tag{75}$$

and $\mathscr{L}$ is a dimensionless Lagrangian spectrum defined by

$$\mathscr{L}(k, w) = \mathscr{E}(k, w)\mathscr{E}(k, 0)^{-1} \tag{76}$$

where

$$\mathscr{E}(k, w) = \tfrac{1}{2}a(k)\int_0^\infty dD\; a(\mathbf{D})\mathscr{B}(kD) \cdot \langle \mathbf{u}(\mathbf{X}, s) \cdot \mathbf{u}(\mathbf{X} + \mathbf{D}, s\,|\,w)\rangle \tag{77}$$

Note that $\mathscr{E}(k, 0) = E(k)$. See Figure 7 for a graph of $\mathscr{F}(\theta)$, when $N = 3$.

4.3. (*Enstrophy*) *Inertia-Convective Subrange*

In the enstrophy-cascading inertial subrange of two-dimensional turbulence, characterized by the time scale $\lambda^{-1/3}$ and the energy spectrum (72), the dimensionless function $\mathscr{L}(k, w)$ must be independent of k:

$$\mathscr{L} = \mathscr{L}(w\lambda^{1/3}) \tag{78}$$

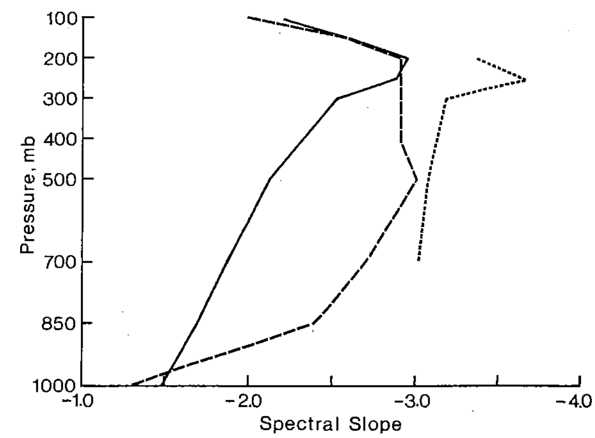

Fig. 6. Slopes of straight line fits to observations of log $E(n)$ versus log n in the atmosphere, for the zonal wave number range $14 \le n \le 25$, from *Baer* [1972] (dotted line), *Chen and Wiin-Nielsen* [1978] (dashed line), and *Boer and Shepherd* [1983] (solid line) [after *Boer and Shepherd*, 1983].

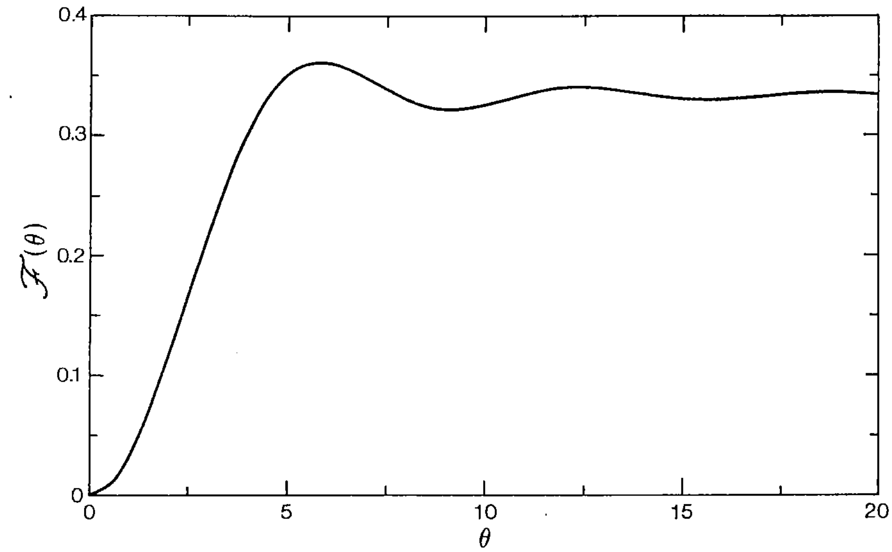

Fig. 7. High-pass filter $\mathcal{F}(\theta)$ relating kinetic energy at wave number k to relative diffusivity at separation R, where $\theta = kR$, in three-dimensional isotropic turbulence. See equation (74).

Combining (72), (73), and (78) yields

$$\eta = 4 \int_s^t \mathcal{L}[(w - s)\lambda^{1/3}]\, dw \int_0^\infty K_r \lambda^{2/3} k^{-3} \mathcal{F}(kR)\, dk \qquad (79)$$

However, the limiting forms of $\mathcal{F}$ given in (75) imply that the integral over k, while convergent as $k \to \infty$, is divergent as $k \to 0$. Since the enstrophy spectrum $k^2E(k)$ must be integrable, it follows that $E(k)$ must be overestimated by (72) at the very low wave numbers where enstrophy is being injected. Thus

$$\int_0^\infty E(k)\mathcal{F}(kR)\, dk \propto R^2 \int_0^\infty k^2 E(k)\, dk \propto R^2 \lambda^{2/3} \qquad (80)$$

leading to

$$\eta \cong bR^2\lambda^{2/3}(t - s) \qquad \lambda^{1/3}(t - s) \to 0 \qquad (81a)$$

$$\eta \cong bR^2\lambda^{1/3} \qquad \lambda^{1/3}(t - s) \to \infty \qquad (81b)$$

In (81), b stands for different dimensionless constants. This convention will be used hereinafter. It is easily seen that (81) holds for any subrange in which the energy spectrum is proportional to k^{-n} when $n \geq 3$. [*Bennett*, 1984; *Babiano et al.*, 1985]. Thus atmospheric observations [*Morel and Larcheveque*, 1974] and oceanic observations (J. F. Price as cited by *McWilliams et al.* [1983]) (see Figures 8 and 9 here), which support (81), are not necessarily indicative of an enstrophy cascade. (Note that if $\eta \propto R^q$ for some power q, then the relative diffusivity ζ in (54) is also proportional to R^q.)

According to (81), dispersion is very slow at first, so it is reasonable to solve the initial value problem of (63) and (64) for $N = 2$, using the large-time estimate (equation (81*b*)) for η, especially as it is the equilibrium variance spectrum

$$F(k) = \lim_{t \to \infty} F(k, t) \qquad (82)$$

which is of interest. The solution is a lognormal distribution for R [*Lundgren*, 1981]:

$$2\pi RP(R, s \mid r, t) = (4\pi\sigma R^2)^{-1/2} \exp\left[-(L - 2\sigma)^2/(4\sigma)\right] \qquad (83)$$

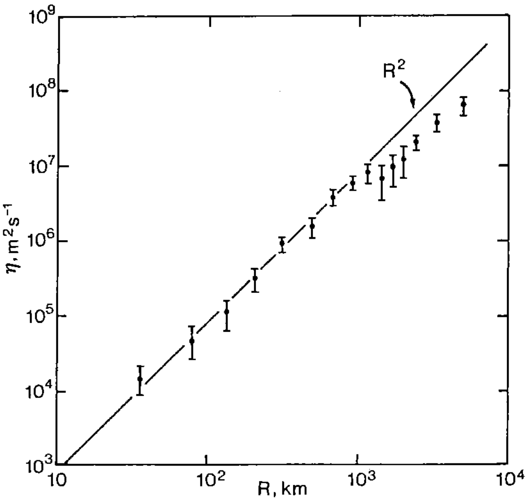

Fig. 8. Relative diffusivity η as a function of separation R, inferred from observations of high-altitude balloon pairs in the southern hemisphere. The straight line has a slope of $+2$. [after *Morel and Larcheveque*, 1974].

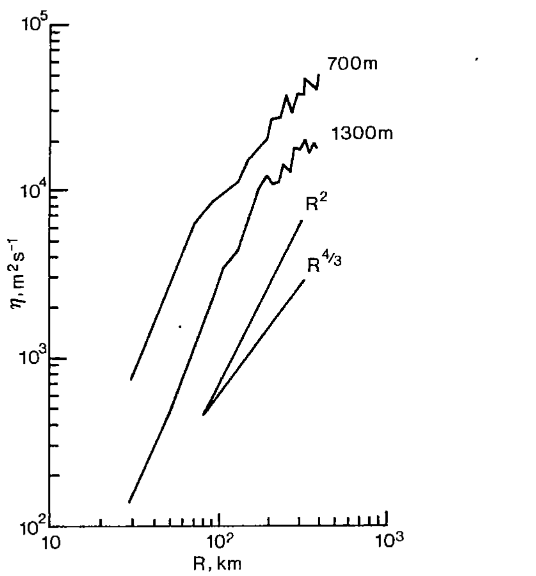

Fig. 9. Relative diffusivity η as a function of separation R, inferred from observations of subsurface ocean drifters at depths of 100 m and 1300 m, at the southern edge of the Gulf Stream recirculation gyre. The straight lines have slopes of $+2$ and $+\frac{4}{3}$ (after J. F. Price, cited by *McWilliams et al.* [1983]).

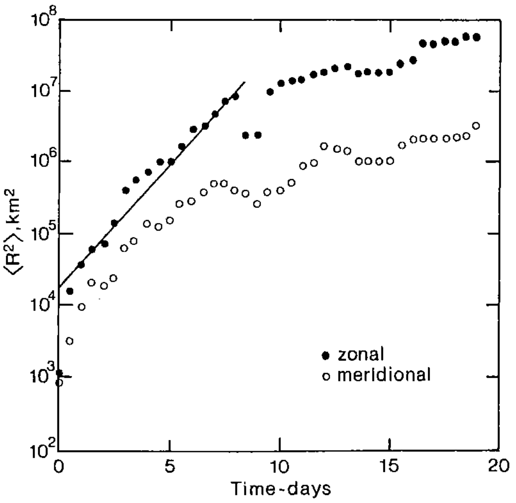

Fig. 10. Separation variance $\langle R^2 \rangle$ as a function of time from launch, for high-altitude balloons in southern hemisphere subtropics. The straight line indicates exponential growth [after *Er-El and Peskin*, 1981].

where $L = \ln (R/r)$ and $\sigma = (t - s)\lambda^{1/3}$. It follows immediately that

$$\langle R^n \rangle = r^n e^{n(n+2)\sigma} \qquad -\infty < n < \infty \tag{84}$$

There is large-scale atmospheric evidence in support of (84) for $n = 2$, [*Er-El and Peskin*, 1981] (see Figure 10). Again, while these observations are consistent with a relative diffusivity $\zeta \propto R^2\tau^{-1}$ where τ is some time scale such as $\lambda^{-1/3}$ [*Lin*, 1972], they only imply that $E(k) \propto k^{-q}$, for some $q \geq 3$.

In order to calculate the equilibrium variance spectrum $F(k)$ using (23), (82), and (83), it is convenient to interchange orders of integration and then use the result that for P given by (83),

$$\lim_{t\to\infty} \int_0^t P(R, s\,|\,r, t)\, ds = \lambda^{-1/3}(4\pi r^2)^{-1} \qquad R \leq r \tag{85a}$$

$$\lim_{t\to\infty} \int_0^t P(R, s\,|\,r, t)\, ds = \lambda^{-1/3}(4\pi R^2)^{-1} \qquad R > r \tag{85b}$$

The time-integrated distribution (equation (85)) is not normalized; this is to be expected since P is normalized for each s (see (22)). However, given (85), the Fourier integrals in (23) are convergent, so the interchange of orders of integration is justified. Using the identity (equation (17)) then yields [*Bennett and Denman*, 1985]

$$F(k) = b\chi\lambda^{-1/3}kl^{-2} \qquad k < l \tag{86a}$$

$$F(k) = b\chi\lambda^{-1/3}k^{-1} \qquad k > l \tag{86b}$$

This spectral shape, $O(k^{-1})$, is neither red nor blue; every wave number decade makes the same contribution to the total variance. The total is infinite as might be expected as $t \to \infty$, given that there is a stationary source of scalar variance and that scalar diffusion has been ignored. It must be conceded that the result (equation (86b)) could have been deduced using dimensional analysis alone, without having to determine the separation pdf (equation (83)) or having to evaluate the integral (equation (23)). We shall return to this important point later.

4.4. (*Energy*) *Inertia-Convective Subrange*

Consider the three-dimensional energy-cascading subrange characterized by energy dissipation rate ε and Eulerian energy spectrum (70). On dimensional grounds the dimensionless Lagrangian spectrum $\mathscr{L}(k, w)$ must be of the form [*Kraichnan*, 1966]

$$\mathscr{L}(k, w) = \mathscr{L}(wk^{2/3}\varepsilon^{1/3}) \tag{87}$$

Combining (70), (73), and (87) yields

$$\eta = 4 \int_0^\infty dk\; K_V \varepsilon^{2/3} k^{-5/3} \mathscr{F}(kR) \int_s^t dw\; \mathscr{L}[(w - s)k^{2/3}\varepsilon^{1/3}] \tag{88}$$

By definition, $\mathscr{L}(0) = 1$, and it will be assumed that $\mathscr{L}$ is integrable from 0 to ∞. Then the limiting forms of $\mathscr{F}$ given in (75) imply that the integral over k in (88) is convergent as $k \to 0$ and as $k \to \infty$. Consequently [*Kraichnan*, 1966],

$$\eta \cong b(t - s)\varepsilon^{2/3}R^{2/3} \qquad (t - s)\varepsilon^{1/3}R^{-2/3} \to 0 \tag{89a}$$

$$\eta \cong b\varepsilon^{1/3}R^{4/3} \qquad (t - s)\varepsilon^{1/3}R^{-2/3} \to \infty \tag{89b}$$

The asymptotic form (equation (89b)) was inferred by *Richardson* [1926] from observations. Seemingly substantial support has been obtained subsequently [e.g., *Okubo*, 1971] (see Figure 11 here) over a very wide range of scales: 10 m $< R <$ 10^9 m(!). However, flow on the larger scales could hardly be described as three-dimensional isotropic turbulence characterized by a well-defined ε. As *Okubo* [1971] points out, diagrams like Figure 11 here are misleading; they are not plots of $D\langle R^2 \rangle/Dt$ against R; rather, they are plots against $\langle R^2 \rangle^{1/2}$. Thus all the "$\frac{4}{3}$" curve substantiates is the cubic time dependence: $\langle R^2 \rangle \propto t^3$. The latter is also characteristic of particle pairs taking independent random walks in a shear flow (see

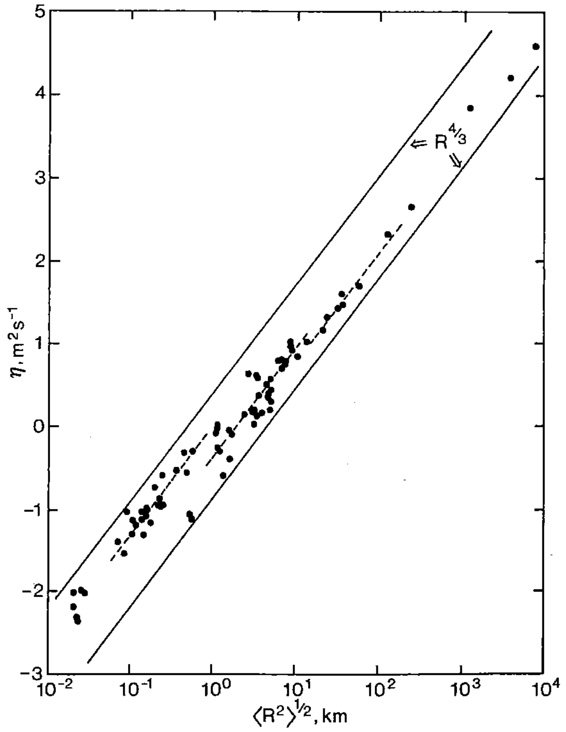

Fig. 11. Relative diffusivity η as a function of rms separation $\langle R^2 \rangle^{1/2}$, inferred from observations at the ocean surface obtained in various experiments. The straight lines have the slope $+\frac{4}{3}$; this merely indicates $\langle R^2 \rangle \propto t^3$ [after *Okubo*, 1971].

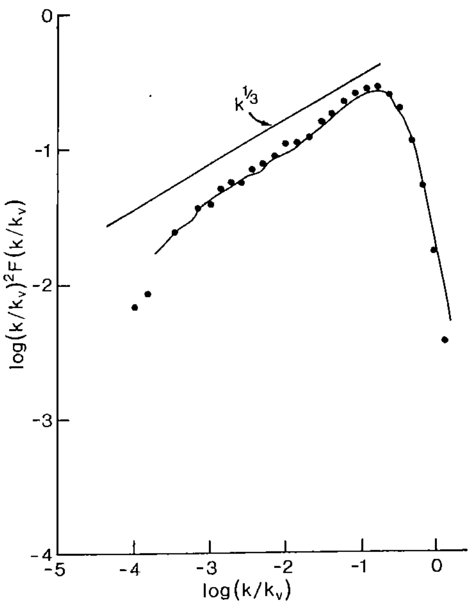

Fig. 12. Temperature variance dissipation spectrum $(k/k_v)^2F(k/k_v)$ as a function of k/k_v, in an atmospheric boundary layer in Minnesota. The straight line has a slope of $+\frac{1}{3}$ [after *Champagne et al.*, 1977].

Bowden [1965] or Appendix I here). In conclusion, it is doubtful that there is any proper, direct observational evidence for (89*b*), although recent laboratory measurements by *Mory and Hopfinger* [1986] are suggestive. Nevertheless, we shall pursue its consequences here.

Again, it is argued that (89*a*) indicates very slow dispersion at first, so it is reasonable to solve the initial value problem (equations (63) and (64)) for $N = 3$, using the large-time estimate (equation (89*b*)) for η. The solution is

$$P(R, s\,|\,r, t) = (8\pi\theta R^{7/6}r^{7/6})^{-1} \cdot \exp\left[-9(r^{2/3}+R^{2/3})(4\theta)^{-1}\right] I_{7/2}[9r^{1/3}R^{1/3}(2\theta)^{-1}] \quad (90)$$

where $\theta = b\varepsilon^{1/3}(t-s) > 0$, and $I_{7/2}$ is a modified Bessel function of the first kind. It is readily shown using (90) that $\langle R^2\rangle = b\varepsilon^{2/3}(t-s)^3$. In the limit $R^{-2/3}\theta \to 0$ but $R \gg r$, (90) has the asymptotic form

$$P(R, s\,|\,r, t) \sim [8\pi(\tfrac{9}{2})!]^{-1}(\tfrac{9}{2})^{7/2}\theta^{-9/2} \exp\left[-9R^{2/3}(4\theta)^{-1}\right] \quad (91)$$

which is the self-similar solution of *Richardson* [1926], independent of initial separation r. As such, it is not useful for the evaluation of F using (23). However, all that is required of (90) is

$$\lim_{t\to\infty}\int_0^t P(R, s\,|\,r, t)\, ds = b\varepsilon^{-1/3}r^{-7/3} \qquad R < r \quad (92a)$$

$$\lim_{t\to\infty}\int_0^t P(R, s\,|\,r, t)\, ds = b\varepsilon^{-1/3}R^{-7/3} \qquad R > r \quad (92b)$$

Evaluation of the Fourier integrals in (23) is not as tidy as for (85), and it is clearest to proceed in stages. First, we calculate the scalar field covariance at equilibrium:

$$V(r) = \lim_{t\to\infty}\langle C(\mathbf{x}+\mathbf{r}, t)C(\mathbf{x}, t)\rangle \quad (93)$$

which is related to $F(k)$ by

$$F(k) = a(k)\int_0^\infty dr\; a(r)\mathscr{B}(kr)V(r) \quad (94)$$

That is, we defer the Fourier integral over r in (23). The approximate result

$$V(r) \cong b\chi\varepsilon^{-1/3}(l^{-2/3} - b'r^{2/3}) \quad (95)$$

holds for $r \ll l^{-1}$. Note that the total scalar variance is found to have a finite equilibrium value:

$$V(0) \cong b\chi\varepsilon^{-1/3}l^{-2/3} \quad (96)$$

even though scalar dissipation has been neglected. On the other hand, it will be seen that the total scalar dissipation rate has an infinite value rather than the correct value χ. Note also the scalar structure function:

$$\lim_{t\to\infty}\langle[C(\mathbf{x}+\mathbf{r}, t) - C(\mathbf{x}, t)]^2\rangle = 2V(0) - 2V(r) \cong b\chi\varepsilon^{-1/3}r^{2/3} \quad (97)$$

Monin and Yaglom [1975] review substantial evidence in support of (97), over a wide range of scales. The scalar spectrum $F(k)$ may be determined from (97) using the inverse integral transform [*Batchelor*, 1960, p. 123]

$$2V(0) - 2V(r) = 2(2\pi)^{-N}\int_0^\infty F(k)[1 - \mathscr{B}(kr)]\, dk \quad (98)$$

It may be seen by inspection that the solution of (98), given (97), is

$$F(k) \cong b\chi\varepsilon^{-1/3}k^{-5/3} \quad (99)$$

In particular, the integral in (98) is convergent both at $k = 0$ and $k = \infty$. There is a wealth of atmospheric and oceanic data in support of (99). *Gargett* [1985] reviews the literature and presents some new high-quality data (see Figures 12 and 13 here).

The result (equation (99) was originally obtained using dimensionless arguments alone, by *Obhukov* [1949], *Corrsin* [1951], and *Batchelor* [1959]. Detailed calculations as above would seem unjustified, especially as dimensional arguments were used to infer the shape of $E(k)$ and hence η. The justification is that the success of the detailed calculations provides support for (89*b*), Richardson's $\frac{4}{3}$ law. *Batchelor* [1952] argued, to the contrary, that η should be independent of R; on dimensional grounds this implies

$$\eta = b\varepsilon(t-s)^2 \quad (100)$$

It follows that the diffusivity tensor η_{ij} must be just $\eta\delta_{ij}$. Then (49) for the vector separation pdf is readily solved, yielding an uncorrelated multivariate normal distribution for $\mathbf{R}$, with zero mean and variance

$$\langle R^2\rangle = b\varepsilon^{1/3}(t-s)^3 \quad (101)$$

which is well supported by data, as already mentioned. Combining this result with (19) leads to the equilibrium scalar variance spectrum

$$F(k) = b\chi\varepsilon^{-1/3}l^{-2/3}\delta(k - l) \quad (102)$$

indicating no cascade of scalar variance. Note that $\delta(k-l)$ has the same dimensions as k^{-1}.

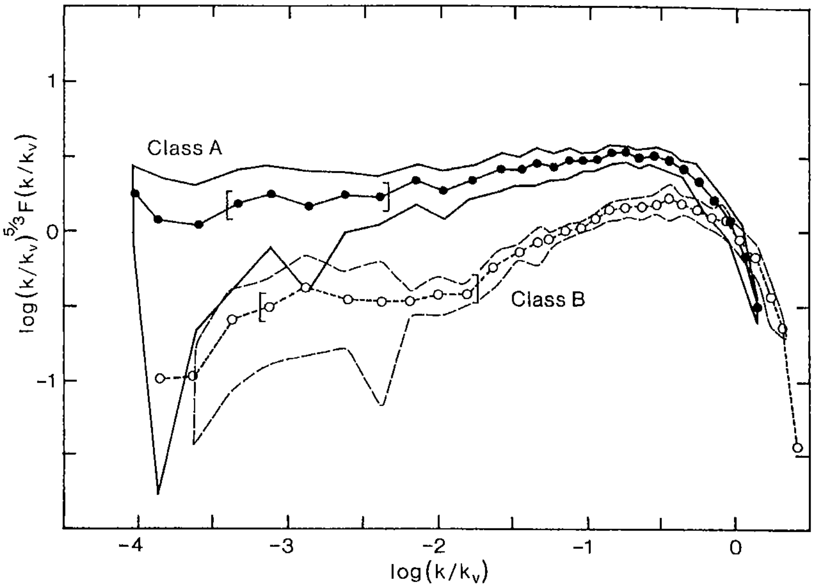

Fig. 13. Class-average, $\frac{5}{3}$-moment temperature spectra $(k/k_V)^{5/3}F(k/k_V)$ in a British Columbia fjord, as functions of k/k_V. Class A data pass stringent tests for statistical isotropy and have high signal-to-noise ratios. Class B data depart from isotropy and have lower signal-to-noise ratios. The envelopes indicate the variance within the classes. The approximately level segments between the brackets indicate $F(k) \propto k^{-5/3}$ [after *Gargett*, 1985].

4.5. *Viscous-Convective Subrange*

The inertia-convective subrange discussed in the previous subsection involves wave numbers much less than the Kolmogorov value k_V. In order to compute the variance spectrum at wave numbers much greater than k_V, it is necessary to determine $P(R, s \mid r, t)$ for values of $R \ll k_V^{-1}$. In particular, the diffusivity $\eta(R, s \mid t)$ is needed for $R \ll k_V^{-1}$. The inertial subrange formula (equation (88)) may still be used, provided the upper limit of integration over k is set at the cutoff fk_V (see (71)), and $\mathscr{F}(kR)$ may be replaced by bk^2R^2. It follows that

$$\eta \cong b\Omega^2(t-s)R^2 \qquad \Omega(t-s) \to 0 \qquad (103a)$$

$$\eta \cong b\Omega R^2 \qquad \Omega(t-s) \to \infty \qquad (103b)$$

where $\Omega = \varepsilon^{1/3}k_V^{2/3} = (\varepsilon/\nu)^{1/2}$ is, to within a dimensionless constant, the rms vorticity.

The approximate diffusivity (equation (103)) is of the same form as that in the enstrophy inertia-convective subrange (equation (81)). We may immediately infer from (86) that the scalar variance spectrum is

$$F(k) = b\chi\Omega^{-1}kl^{-2} \qquad k < l \qquad (104a)$$

$$F(k) = b\chi\Omega^{-1}k^{-1} \qquad k > l \qquad (104b)$$

This result was originally obtained by *Batchelor* [1959], again by *Kraichnan* [1974], and also by *Lesieur et al.* [1981]. There is no clear supporting evidence for (104) atmospheric or oceanic data. Convective subranges exist only where the scalar diffusion rate κk^2 is much smaller than the strain rate $(\varepsilon/\nu)^{1/2}$, that is, $k \ll k_B$ where $k_B = k_V\ Pr^{1/2}$ is the Batchelor wave number and $Pr = \nu/\kappa$ is the Prandtl number. Thus viscous-convective subranges require $k_V \ll k \ll k_V\ Pr^{1/2}$, or $1 \ll Pr^{1/2}$. For air, $Pr^{1/2} = 0.85$, while for water, $Pr^{1/2} = 2.6$. However, several of the scalar spectra reported by *Gargett* [1985] show "$-\frac{5}{3}$" inertia-convective ranges which flatten out before rolling off above k_B.

4.6. *Transition*

The energy spectrum $E(k)$ has a well-defined transition from the (energy) inertial to the viscous subrange at $k \cong k_V/10$ (see Figure 5). All that can be inferred thus far for the scalar spectrum is a transition from the $-\frac{5}{3}$ inertial-convective subrange to the "-1" viscous-convective subrange, also at $k \sim k_V/10$. Thus it is a little surprising that observations [e.g., *Gargett*, 1985] show well-defined scalar transitions at $k \cong k_V/100$. It is argued here that no other physics need be involved in order to explain this misalignment; such a large numerical factor is to be expected. A detailed solution of (63), using a coefficient η which interpolates between the $\frac{4}{3}$ law (equation (89b)) and the "2" law (equation (103b)), might reveal such a factor, but at considerable computational effort. The interpolation formula would be arbitrary; that is, it could introduce arbitrary numerical factors. Instead, an examination of the dependence of η on E and $\mathscr{L}$ will itself reveal an appropriate numerical factor.

It is sufficient to examine η in the limit $(t-s) \to \infty$. Let $\mathscr{L}(k, t)$ have the inertial range form (equation (87)). As was stated in the preceding section, the range of integration for k in (88) is effectively limited to $k \le fk_V$; in any case, this choice can only prejudice the result in favor of the inertia-convective subrange form for η (i.e., possibly pushing the scalar transition above k_V). Then we have

$$\eta = b\varepsilon^{1/3} \int_0^{fk_V} R^{-7/3}\mathscr{F}(kR)\, dk \qquad (105)$$

The integral over k in (105) may be evaluated numerically. The results are shown in Figure 14. They indicate a well-defined transition for η at a separation R almost a decade greater than the cutoff length $(fk_V)^{-1}$. This is indicative of a scalar transition in wave number space almost a decade below fk_V. The source of this numerical factor of $\cong 6$ is the slow rise of the high-pass filter $\mathscr{F}(a)$ from $a^2/30$ for $a \ll 1$ to $\frac{1}{3}$ as $a \to$

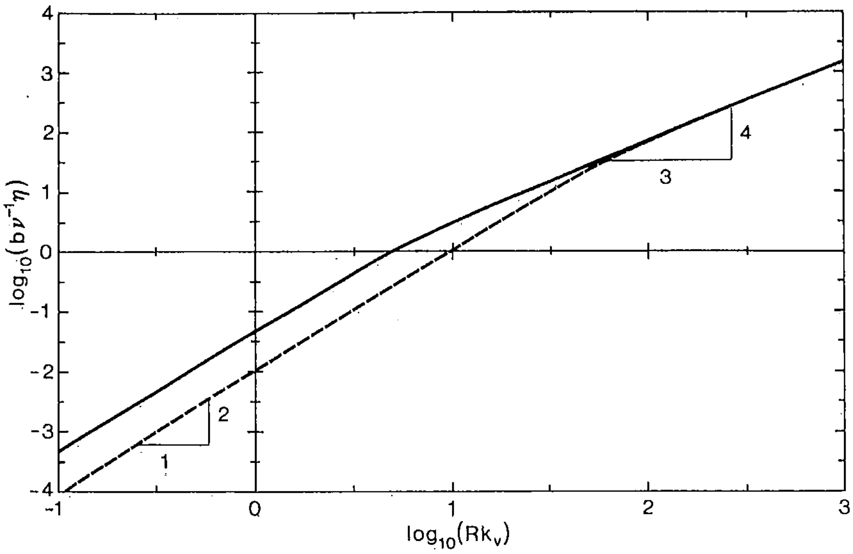

Fig. 14. Large-time relative diffusivity η as a function of separation, according to the truncated model (equation (71)) for the energy spectrum: $f = 1$, and the transition from the R^2 range to the $R^{4/3}$ range is at $R \cong 6k_\nu^{-1}$ (solid line); $f = 0.1$, and the transition is at $R \cong 60k_\nu^{-1}$ (dashed line). That is, the transition occurs at a separation close to an order of magnitude larger than the length scale of maximum dissipation.

∞ (see Figure 7). Note that the first maximum of $\mathscr{F}$ occurs at $a \cong 6$. The shape of $\mathscr{F}$ is purely a consequence of the geometry of isotropic turbulence.

If the Prandtl number $Pr = \nu/k$ is not large, then the viscous-convective subrange should not be well defined. The inertia-convective subrange should have a smooth transition to the rapidly decaying viscous-diffusive subrange described in the next section. However, a clear "bump" in the scalar spectrum is commonly observed for $k = O(k_\nu)$ [*Champagne et al.*, 1977; *Williams and Paulson*, 1977]. It is suggested here that the bump is a latent viscous-convective subrange, which exists because of the tendency of η to convert to the 2 law for $k \ll k_\nu$. Similar conclusions were reached by *Hill* [1978], who calculated spectra corresponding to several models of the spectral transfer rate. Those models include two or three disposable parameters. The analysis here has none.

5. DIFFUSIVE SUBRANGES

So far, scalar diffusion has been neglected; specifically, the diffusion factor $g(\mathbf{k}, t, s)$ appearing in the representation (18) has been replaced by unity. This assumes that the initial diffusion rate κk^2 is much smaller than the turbulent strain rates. The assumption will now be relaxed; approximate forms for g will be devised, and diffusive subrange spectra will be calculated.

5.1. *(Enstrophy) Inertia-Diffusive Subrange*

Consider the enstrophy-cascading inertial subrange of two-dimensional turbulence. It will be seen in section 5.3 that the rate of infinitesimal line stretching is characterized by the rms strain rate and so is $O(\lambda^{1/3})$ here, where λ is the enstrophy cascade rate. The logarithmic separation rate for particle pairs, defined by $R^{-2}\eta$, is also $O(\lambda^{1/3})$ according to (81*b*). These rates are greatly exceeded by the scalar diffusion rate κk^2 if $(\lambda\kappa^{-3})^{1/6} \ll k$. On the other hand, k must be less than the upper limit of the enstrophy-cascading inertial subrange of the turbulence.

In this subrange, infinitesimal line stretching is negligible, so the diffusion factor $g(\mathbf{k}, t, s)$ may be approximated by exp $[-\kappa k^2(t - s)]$. In particular, the fundamental assumption of spatially uniform stretching factors is vindicated here; these factors do not depart significantly from unity during the diffusion process. Only the marginal statistics of separation are required, and the short-time relative diffusivity (equation (89*a*)) should be appropriate. Indeed, as a first approximation it would seem sufficient to approximate the separation pdf $P(R, s\,|\,r, t)$ by its initial form $a(R)^{-1}\delta(R - r)$. However, such an approximation leads immediately to the scalar variance spectrum

$$F(k) = b\chi a(k)\delta(k - l)(\kappa k^2)^{-1} \tag{106}$$

where $a(k) = 2\pi k$ for two dimensions. That is, there is no cascade from the injection wave number l, as a consequence of having entirely ignored relative diffusion. Clearly, it is necessary to recognize that P has a small but finite spread about $R = r$. For example, the variance is $O[\lambda^{2/3}(t - s)^2 r^2]$. A correction to (106) may be calculated this way, but the result depends upon the injection wave number l. That is, there is no universal form for the inertia-diffusive subrange here.

The model used above (and in all of this article so far) assumes that the scalar field is sustained by an isotropic source. This is an idealization. A more realistic model (and one which is far more easily realized in practice) has no external source of scalar variance but instead has a mean scalar concentration with a gradient. In particular, it will be assumed that the gradient is uniform in space and time. Without loss of generality it will be assumed that the gradient is parallel to one of the space axes:

$$\nabla\langle C\rangle = (\Gamma, 0, 0) \tag{107}$$

In this model the turbulent velocity field is still assumed to be stationary, isotropic, and with zero mean. Fluctuations in C will be induced by turbulent advection of the mean scalar gradient or possibly by random initial values for C, but this latter possibility will be ignored by assuming $C' \equiv 0$ at $t = 0$. The prime denotes a fluctuation in C.

It is easily seen that C' satisfies the advection-diffusion equation (equation (1)) with the random source S replaced by $-u_1(\mathbf{x}, t)\Gamma$, which is the advection down the mean scalar gradient. Hence the solution is, according to the representation (5),

$$C'(\mathbf{x}, t) = -\Gamma \int_0^t ds \int d\mathbf{y}\, G(\mathbf{x} - \mathbf{y}, t, s) u_1(\mathbf{y}, t \,|\, s) \tag{108}$$

It is now straightforward to derive an expression for the equilibrium scalar variance spectrum $F(k)$. Again, the inertia-diffusive subrange form will be assumed for the diffusion factor: $g(\mathbf{k}, t, s) \cong \exp[-2\kappa k^2(t - s)]$. The expression for $F(k)$ includes the Lagrangian velocity correlation $\langle \mathbf{u}(\mathbf{y}_1, t\,|\,s_1) \cdot \mathbf{u}(\mathbf{y}_2, t\,|\,s_2)\rangle$ where $0 \le s_1$ and $s_2 \le t$. However, we need only consider $(t - s_{1,2}) = O(\kappa k^2)^{-1}$, which is much less than the decorrelation time scales of the turbulence (here $O(\lambda^{-1/3})$). As a result, the Lagrangian velocity correlation may be approximated by the Eulerian covariance $\langle \mathbf{u}(\mathbf{y}_1, t\,|\,t) \cdot \mathbf{u}(\mathbf{y}_1, t\,|\,t)\rangle = \langle \mathbf{u}(\mathbf{y}_1, t) \cdot \mathbf{u}(\mathbf{y}_2, t)\rangle = V_{uu}(\mathbf{y}_1 - \mathbf{y}_2)$ for stationary homogeneous turbulence. The latter is related to the kinetic energy spectrum by the Fourier transform (equation (69)). With these approximations the expression for $F(k)$ may be evaluated analytically to yield

$$F(k) = b\Gamma^2 E(k)(\kappa k^2)^{-2} \tag{109}$$

In the enstrophy subrange, $E(k) = b\lambda^{2/3}k^{-3}$, so

$$F(k) = b\Gamma^2\lambda^{2/3}\kappa^{-2}k^{-7} \tag{110}$$

5.2. *(Energy) Inertia-Diffusive Subrange*

In the energy-cascading inertial subrange, infinitesimal line stretching proceeds at the rms strain rate $\Omega = (\varepsilon/\nu)^{1/2}$, while particle pairs separate at the rate $\varepsilon^{1/3}k^{2/3}$ (if we consider separations $r \sim k^{-1}$). Let $k_D = \varepsilon^{1/4}\kappa^{-3/4} = k_V\, Pr^{3/4}$. If $Pr \ll 1$, then there is an inertia-diffusive subrange, $k_D \ll k \ll k_V$, between the inertial-convective and viscous-diffusive subranges. In this intermediate range, $(\varepsilon/\nu)^{1/2} \ll \varepsilon^{1/3}k^{2/3} \ll \kappa k^2$. That is, the molecular diffusion rate greatly exceeds the local separation rate, which in turn exceeds the infinitesimal stretching rate. In particular, the diffusion factor $g(\mathbf{k}, t, s)$ has the simple form given previously, and only marginal statistics of separation are needed to compute $F(k)$.

The isotropic source model leads to a scalar variance spectrum dependent upon the injection wave number l; that is, a universal form is not found. The uniform gradient model leading to (109), combined with the energy subrange (equation (70)), yields

$$F(k) = b\Gamma^2\varepsilon^{2/3}\kappa^2 k^{-17/3} \tag{111}$$

This result is originally owing to *Batchelor et al.* [1959]. It was also derived by *Kraichnan* [1968] using the Lagrangian history direct interaction approximation. More recently *Lesieur et al.* [1981] and *Lesieur and Herring* [1985] have derived (110) and (111) using eddy-damped quasi-normal closure theory.

It may be noted that all these other derivations assume an isotropic source model but in effect argue that a low wave number component of the scalar field is in practice indistinguishable from a mean gradient in the field. They effectively identify the (squared) mean gradient Γ^2 with the (scaled) source strength $\chi\kappa^{-1}$. What is omitted in these other derivations is the variance production term $\langle CS\rangle$. It is claimed here that this term is a source of nonuniversality. The observations of *Clay* [1973] in a laboratory channel using mercury ($Pr = 0.02$) clearly support (111), but the experiment is best described by the mean gradient model. It seems improbable that an isotropic source can be devised, so this claim will be difficult to test.

5.3. *Viscous-Diffusive Subrange*

As was discussed in section 4.5, there is a viscous-convective subrange, $k_V \ll k \ll k_B = k_V\, Pr^{1/2}$, provided the Prandtl number Pr is very large. Beyond this range, that is, for $k \gg k_B$, scalar diffusion cannot be neglected. There will be fluctuations in the diffusion factor $g(\mathbf{k}, t, s)$ owing to infinitesimal line stretching; this proceeds at the local or rms strain rate $\Omega = (\varepsilon/\nu)^{1/2}$, as does (logarithmic) particle pair separation (see (103*b*)). Thus joint statistics of stretching and separation are required in order to estimate $F(k)$ using (18). Specifically, what are required are the joint statistics of finite separation $\mathbf{R}$ and the infinitesimal displacement $\delta\mathbf{A}$ appearing in (6).

To this end it is convenient to introduce the vector $\mathbf{h}$ where

$$h_i = k_j \frac{\partial A_j}{\partial x_i}[\mathbf{A}(\mathbf{x}, t\,|\,s), s\,|\,t] \tag{112}$$

Then $\mathbf{k} \cdot \mathbf{Q} \cdot \mathbf{k} = \mathbf{h} \cdot \mathbf{h} = h^2$. It is shown in Appendix II that

$$\partial\mathbf{h}/\partial s = -\mathbf{W}^T\mathbf{h} \tag{113}$$

subject to $\mathbf{h} = \mathbf{k}$ at $s = t$, where

$$W_{ij} = \frac{\partial u_i}{\partial x_j}(\mathbf{x}, t\,|\,s) \tag{114}$$

Meanwhile, $\mathbf{R}$ satisfies

$$\partial\mathbf{R}/\partial s = \mathbf{u}(\mathbf{X} + \mathbf{R}, s) - \mathbf{u}(\mathbf{X}, s) \tag{115}$$

subject to $\mathbf{R} = \mathbf{r}$ at $s = t$. For $R \ll k_V^{-1}$, separation is controlled in the rms sense by eddies of scale $k^{-1} = k_V^{-1} \gg R$, so $\mathbf{R}$ obeys, essentially,

$$\partial\mathbf{R}/\partial s \simeq \mathbf{W}\mathbf{R} \tag{116}$$

It may be noted that $\mathbf{R}$ and $\mathbf{h}$ are dual vectors in the sense that $\mathbf{h} \cdot \mathbf{R} \equiv \mathbf{k} \cdot \mathbf{r}$, for all s. However, there is no obvious statistical relationship between the separation $\mathbf{R}$ and the vector $\mathbf{L}$ appearing in (6). We shall simply neglect that "drift" term; as a result, we have

$$\partial g/\partial s = \kappa h^2 g \tag{117}$$

subject to $g = 1$ as $s = t$. This system of random differential equations (equations (113), (116), and (117)) will provide the required joint statistics of separation and stretching.

It is shown in Appendix II that the calculation of the scalar variance spectrum reduces to

$$F(k, t) = \chi \int da(\mathbf{k}) \int da(\mathbf{l}) \int_0^t ds \int dg\, P(g, \mathbf{k}, t\,|\,1, \mathbf{l}, s) g^2 \tag{118}$$

where P is the joint pdf for the system

$$\partial g/\partial t = -\kappa k^2 g \tag{119a}$$

$$\partial \mathbf{k}/\partial t = -\mathbf{W}^T\mathbf{k} \tag{119b}$$

with $g = 1$ and $\mathbf{k} = \mathbf{l}$ at $t = s$. *Kraichnan* [1974] arrived at this point by an analogous construction. He proposed a one-dimensional model for $k = |\mathbf{k}|$:

$$\partial k/\partial t = \alpha k \tag{120}$$

with $k = l$ at $t = s$. The statistically stationary, random strain rate $\alpha(t)$ has a positive mean and a white noise variance v^2. That is, (120) should be written as

$$dk = \langle\alpha\rangle k\, dt + 2^{1/2} v k\, d\omega \tag{121}$$

where $\omega(t)$ is the Wiener process of unit variance [*van Kampen*, 1981]. Both $\langle\alpha\rangle$ and v^2 are $O(\Omega)$. Kraichnan argued that $\langle\alpha\rangle = Nv^2$ where N is the number of space dimensions in (119*b*). Once the joint pdf $P(g, k, t\,|\,1, l, s)$ for the system (119*a*) and (121) has been found, the equilibrium spectrum is given by

$$F(k) = \chi \int_0^\infty dt \int_0^1 dg\, P(g, k, t\,|\,1, l, 0) g^2 \tag{122}$$

The joint pdf in (122) satisfies the Fokker-Planck equation

$$P_t = -\langle\alpha\rangle(kP)_k + \kappa k^2(gP)_g + v^2 k(kP_k)_k \tag{123}$$

in the Stratonovitch interpretation, for which α is regarded as a process with a vanishingly small decorrelation time [*van Kampen*, 1981]. The initial condition is

$$P(g, k, 0\,|\,1, l, 0) = \delta(g-1)\delta(k-l) \tag{124}$$

Steady state and time-dependent solutions have been obtained by *Kraichnan* [1974] and *Bennett* [1986], respectively. The solution is particularly simple in the case $v = 0$, which corresponds to *Batchelor*'s [1959] uniform strain model. The spectrum is

$$F(k) = \chi\langle\alpha\rangle^{-1}k^{-1} \exp\left[-\kappa\langle\alpha\rangle^{-1}(k^2 - l^2)\right] \tag{125}$$

as given by Batchelor. If $v > 0$, then the spectrum is of the form

$$F(k) \propto k^w \exp\left[-(2\kappa v^{-1})^{1/2} k\right] \tag{126}$$

as $k \to \infty$ and $l \to 0$ [*Kraichnan*, 1974], where $w = \frac{1}{2}(\langle\alpha\rangle v^{-1} - 3)$. However, if the range of integration over g in (122) is restricted to $g_0 \le g \le 1$ where $0 < g_0$, then, still for the case $v > 0$, the spectrum is asymptotically of the form

$$F(k) \propto k^{2w} f_0^{\,w-1/2} \exp\left[-\kappa\langle\alpha\rangle^{-1}(4vf_0)^{-1}k^2\right] \tag{127}$$

where $f_0 = \ln g_0$. This is essentially of the same form as the Batchelor spectrum (equation (125)) even though the strain rate α is now random. The cutoff g_0 may represent the threshold sensitivity of a measuring instrument.

The adoption of a white noise model for the strain rate $\alpha(t)$ as in (121) is extreme. Consider instead the model

$$dk/dt = k\Omega M(\theta) \tag{128}$$

where θ is a standard normal random variable and M is some positive-valued functional form. It is assumed that θ is independent of time or else is a stationary process with a very long decorrelation time. The spectrum $F(k, \theta)$ for a particular realization of θ is given by the Batchelor form (equation (125)), with $\langle\alpha\rangle$ replaced by $\Omega M(\theta)$. The spectrum $F(k)$ is obtained by averaging over θ. Asymptotic forms for $F(k)$ may be obtained using the method of steepest descent, as $k \to \infty$. There are several interesting examples:

$$M(\theta) = e^\theta$$

This lognormal strain rate is a natural choice. The spectrum is

$$F(k) \propto \chi\Omega^{-1}k^{-2} \ln(k/k_B) \exp\left[-\tfrac{1}{2}\ln^2(k/k_B)\right] \tag{129}$$

Note that $k_B{}^2 = \Omega\kappa^{-1}$. Thus intermittency of the strain rate leads to a very broad spectrum.

$$M(\theta) = O(1) \qquad |\theta| \to \infty$$
$$M'(\theta) = O(1) \qquad |\theta| \to \infty$$

The spectrum is, asymptotically as $k \to \infty$,

$$F(k) \propto \chi\Omega^{-1}k^{-1} \exp\left[-b_1(k/k_B)^2 - b_2(k/k_B)^4\right] \tag{130}$$

where b_1 and b_2 are positive, bounded dimensionless functions of k/k_B. This resembles the Batchelor form (equation (125)).

It is instructive to compare this array of results (equations (125), (126), (127), (129), and (130)) with observations. *Gargett* [1985] provides a review and presents high-quality data from a turbulent coastal channel (see Figure 15). These recent observations support the Batchelor spectrum (equation (125)), but no universal value is found for $q = \Omega\langle\alpha\rangle^{-1}$. (In terms of q the exponential in (125) becomes $\exp\left[-q(k^2 - l^2)k_B{}^{-2}\right]$). For large signal-to-noise ratios (small g_0), Gargett finds large values for q; this contradicts (127). The most plausible model is (130); if the spectrum were of this form, then fitting the Batchelor form to the data (i.e., assuming $b_2 \equiv 0$) would lead to an overestimate for b_1. The safest conclusion is that while theoretical models of the viscous-diffusive subrange are highly sensitive to model details, Batchelorlike spectral forms are ubiquitous, but universality is not likely.

6. TOTAL SCALAR VARIANCE

So far, the wave number spectrum of the scalar field has been examined, given a random isotropic source of scalar variance. In this section the total scalar variance is estimated for a variety of sources.

6.1. *Random Isotropic Source*

Consider the source S introduced in section 2. It has white noise time dependence (equation (16)) and a simple space correlation (equation (16)) corresponding to injection of scalar variance only at wave numbers with magnitudes $|k| = l$. The total scalar variance may be estimated by integrating the several subranges of the variance spectra.

For the enstrophy inertia-convective range (equation (86)), the result is

$$V_{IC} = b\chi\lambda^{-1/3}\{\tfrac{1}{2} + \ln(k_C/l)\} \tag{131}$$

where k_C is the upper limit of the subrange. If this convective subrange is cut off by scalar diffusion with diffusivity κ, then $k_C = (Um\kappa^{-1})^{1/2}$ where $U = (\varepsilon m^{-1})^{1/3}$ is an rms velocity for the turbulence which has the characteristic length scale m^{-1} described in section 4.1. Assuming $l \simeq m$, that is, scalar variance and kinetic energy are injected at the same wave number,

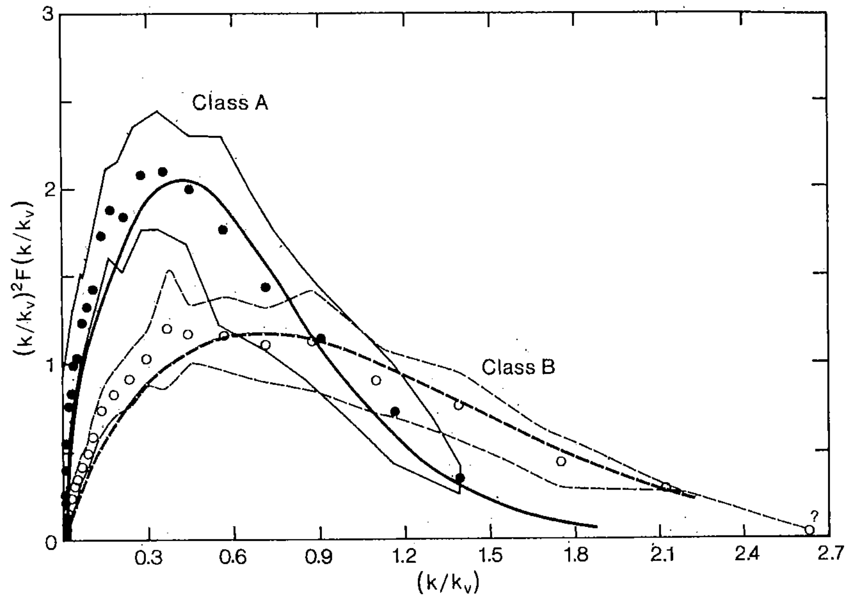

Fig. 15. Variance-preserving plot of temperature variance dissipation spectra in a British Columbia fjord, after *Gargett* [1985]: Batchelor spectrum (equation (125)) with $q \equiv \Omega\langle\alpha\rangle^{-1} = 12$ (heavy solid curve) and Batchelor spectrum with $q = 4$ (heavy dashed curve).

yields $k_C/l \simeq (U\kappa^{-1}m^{-1})^{1/2} = Pe^{1/2}$ where Pe is the Peclet number. Thus

$$V_{IC} \cong b\chi\lambda^{-1/3} \ln Pe \tag{132}$$

However, the detailed physics of the ranges beyond the enstrophy inertia convective are not well defined, so no attempt will be made to estimate their contribution to the total scalar variance. The important point is the dependence, if only logarithmic, of the total variance upon the small-scale parameter k_C.

For the energy inertia-convective subrange the total scalar variance has already been obtained (equation (96)) as the value of the structure function at small separation:

$$V_{IC} \simeq b\chi\varepsilon^{-1/3}l^{-2/3} \tag{133}$$

If $Pr \gg 1$, there is a viscous-convective subrange $k_V \ll k \ll k_V' Pr^{1/2} = k_B$. By virtue of (104*b*), this contributes

$$V_{VC} \simeq b\chi\Omega^{-1} \ln Pr \tag{134}$$

The ratio of the two contributions (equations (133) and (134)) is

$$V_{IC}/V_{VC} = b\ Re^{1/2}(m/l)^{2/3}(\ln Pr)^{-1} \tag{135}$$

where the Reynolds number $Re = Um^{-1}\nu^{-1}$. Thus the inertia-convective contribution should dominate in the geophysical context. The contribution of the viscous-diffusive subrange is negligible.

6.2. *Mean Scalar Gradient*

This state has already been considered in the analysis of the inertia-diffusive subrange (see section 5.1). Again, the mean scalar gradient is

$$\nabla\langle C\rangle = (\Gamma, 0, 0) \tag{136}$$

and scalar fluctuations C' satisfy the advection-diffusion equation (equation (1)) with source $-u_1(\mathbf{x}, t)\Gamma$. Hence C' is given by (108), while the total variance is

$$V = \langle C'^2\rangle = \tfrac{1}{3}\Gamma^2 \int_0^t ds_1 \int_0^t ds_2 \int d\mathbf{y}_1 \int d\mathbf{y}_2\ \langle G(\mathbf{x}-\mathbf{y}_1, t, s_1) \cdot G(\mathbf{x}-\mathbf{y}_2, t, s_2)\mathbf{u}(\mathbf{y}_1, t|s_1)\cdot\mathbf{u}(\mathbf{y}_2, t|s_2)\rangle \tag{137}$$

The physical space diffusion functions are significant only if $|\mathbf{x}-\mathbf{y}_{1,2}|$ are small; they are normalized distribution functions, so in a first approximation,

$$V \simeq \tfrac{1}{3}\Gamma^2 \int_0^t ds_1 \int_0^t ds_2\ \langle\mathbf{u}(\mathbf{x}, t|s_1)\cdot\mathbf{u}(\mathbf{x}+\mathbf{z}, t|s_2)\rangle \tag{138}$$

where $z = |\mathbf{z}|$ is very small, $O(\kappa U^{-1}m^{-1})^{1/2} = m^{-1}\ Pe^{-1/2}$. For isotropic stationary turbulence we have

$$\langle\mathbf{u}(\mathbf{x}, t|s_1)\cdot\mathbf{u}(\mathbf{x}+\mathbf{z}, t|s_2)\rangle = V_{uu}[z, t-\tfrac{1}{2}(s_1+s_2), s_1-s_2] \tag{139}$$

which is a Lagrangian velocity covariance with one space lag and two time lags. In the limit as $t \to \infty$, (138) becomes

$$V \simeq \tfrac{1}{3}\Gamma^2 \int_0^\infty dt_1 \int_0^\infty dt_2\ V_{uu}(z, t_1, t_2) \tag{140}$$

where $t_1 = t - \frac{1}{2}(s_1 + s_2)$ and $t_2 = (s_1 - s_2)$; that is,

$$V \simeq \tfrac{1}{3}\Gamma^2 T \int_0^\infty dt_1\ \bar{V}_{uu}(z, t_1) \tag{141}$$

where $T \simeq (mU)^{-1}$ is an integral time scale for the argument t_2 in V_{uu}, and $\bar{V}_{uu}$ represents the mean of V_{uu} over t_2:

$$\bar{V}_{uu}(z, t_1) = T^{-1}\int_0^\infty dt_2\ V_{uu}(z, t_1, t_2) \tag{142}$$

Since $z \ll m^{-1}$, which is of the order of the space decorrelation scale for $\mathbf{u}$, it will be sufficiently accurate to set $z = 0$ in (142). This does not imply that molecular diffusion has been entirely neglected, since $\bar{V}_{uu}(0, t_1)$ represent a decorrelation of the velocities of two particles which were once very close together (at $s_1 = s_2 = t$; $t_1 = 0$). For a given value of $t_1 \gtrsim T$, their separation should be of the order of $D = (U^2Tt_1)^{1/2}$ since the particles will be making independent random walks (absolute diffusion), and an estimate of the right order magnitude for V should be given by

$$V \simeq b\Gamma^2 T(U^2T)^{-1}\int_0^\infty dD\, V_{uu}(D)D \qquad (143)$$

where $V_{uu}(D)$ is the Eulerian velocity covariance at space lag D:

$$V_{uu}(D) = \langle \mathbf{u}(\mathbf{x}+\mathbf{D}, t)\cdot\mathbf{u}(\mathbf{x}, t)\rangle \qquad (144)$$

That is, since V_{uu} has the length scale $m^{-1} = UT$,

$$V \simeq b\Gamma^2 m^{-2} \qquad (145)$$

provided the integral in (143) is convergent. If it is not convergent, then the velocity field would have such long range correlations that the uniform mean scalar gradient model would not be useful. A possible exception might be gradients on the scale of an oceanic gyre, advected by mesoscale turbulence. However, the question of integrability of a correlation function is not answerable in practice, and the estimate (equation (145)) must bear this stigma. It is bounded but is explicitly independent of the molecular or small-scale diffusivity κ. Nevertheless, it is implicitly dependent upon the existence of a nonzero diffusivity since it is the result of a two-particle calculation (t_1 is the time for which the two particles have been separating after being very close). To explain (145) further, consider the two particles involved in (138). Initially very small, the separation of the two particles will grow as $(\varepsilon t_1{}^3)^{1/2}$ as it crosses the energy inertial subrange. The separation will be of the order of m^{-1} when $t_1 \simeq (m^{-2}\varepsilon^{-1})^{1/3} = m^{-1}U^{-1}$. Each particle will then have been displaced by about $Ut_1 \simeq m^{-1}$; these displacements will be correlated, but subsequent displacements will not; thus (138) yields (145). If the early stage of separation is controlled by an enstrophy inertial subrange for which $\lambda \simeq (Um)^3$, then separation grows as $ze^{\lambda^{1/3}}t_1$, and so displacements will be correlated until $t_1 \simeq m^{-1}U^{-1}|\ln(mz)|$, by which time they are $\simeq Ut_1$. That is, (145) becomes

$$V \simeq b\Gamma^2 m^{-2}\ln Pe \qquad (146)$$

Durbin [1980] also argued for (138). That is, total scalar variance, even in the presence of molecular diffusion, could be calculated with diffusion neglected, by taking the limit of scalar covariance at small space lag. He calculated the displacement correlation (equation (138)) by developing a stochastic model for joint motions of particle pairs. However, he found an unbounded V, growing as $t^{1/2}$ as $t\to\infty$. The reason is that his stochastic model was based on an Eulerian velocity correlation $V_{uu}(D)$ decaying as $D^{-2/3}$ as $D\to\infty$. With this choice the integral in (143) would be divergent. While this decay rate may be appropriate in the energy inertial range, it is probably unrealistically slow for $D \gg m^{-1}$.

6.3. *Random Source of Finite Extent*

It would be attractive to be able to model long-range transport of scalars from localized sources, by tracking particles leaving a point source. For example, let the source strength be given by

$$S(\mathbf{x}, t)\, dt = \chi^{1/2}(2\pi\sigma^2)^{-N/4}e^{-x^2/(4\sigma^2)}\, d\omega(t) \qquad (147)$$

where σ is the radius of the source which is centered at $\mathbf{x} = \mathbf{0}$, N is the dimension of the space, and $\omega(t)$ is the Wiener process. The latter has uncorrelated increments:

$$\langle d\omega(t_1)\, d\omega(t_2)\rangle = \delta(t_1 - t_2)\, dt_1\, dt_2 \qquad (148)$$

Note that the total variance of the total source contribution $\int S\, dt$ is a linear function of time but is independent of the source radius σ:

$$\int \left\langle \int_0^t S(\mathbf{x}, s_1)\, ds_1 \int_0^t S(\mathbf{x}, s_2)\, ds_2 \right\rangle d\mathbf{x} = \chi t \qquad (149)$$

With diffusion neglected ($\kappa = 0$) the solution for the scalar concentration is simply

$$C(\mathbf{x}, t) = \int_0^t ds\, S(\mathbf{x}, t\,|\,s) \qquad (150)$$

An elementary calculation then yields

$$\langle C^2(\mathbf{x}, t)\rangle = \chi\int_0^t ds\, \{2\pi[\sigma^2 + 2(t-s)K]\}^{-N/2}$$
$$\cdot \exp\{-\tfrac{1}{2}|\mathbf{x} + (t-s)\langle\mathbf{u}\rangle|^2/[\sigma^2 + 2(t-s)K]\} \qquad (151)$$

In the derivation of (151) it has been assumed that the particle displacement $\mathbf{A}(\mathbf{x}, t\,|\,s)$ is a multivariate normal random variable, with mean $(t-s)\langle\mathbf{u}\rangle$ and variance $2(t-s)K = 4(t-s)\langle|\mathbf{u}'|^2\rangle T$ where T is the Lagrangian integral time scale of the velocity field. It is also assumed that components of $\mathbf{A}$ are uncorrelated. These assumptions are correct asymptotically as $(t-s)\to\infty$ for isotropic turbulence, as discussed in section 3. If the integral (151) is divergent as $t\to\infty$, then the neglect of scalar molecular diffusion (implied by the use of (150)) is unjustified. Now (151) is convergent, provided that $\langle\mathbf{u}\rangle \neq \mathbf{0}$. Otherwise, it is convergent only for $N \geq 3$. In general, any long-range transport model will involve a mean flow, so convergence is the rule. The next issue is the behavior of the scalar variance as the source radius $\sigma\to 0$. A closed form for (151) is not available, but inferences may be made by examining the integrand at $\sigma = 0$. If $\mathbf{x} \neq \mathbf{0}$, the integral is convergent as $s = 0$. If $\mathbf{x} = \mathbf{0}$, the integral is convergent only for $N = 1$. However, for small values of $(t-s)$ the variance of $\mathbf{A}$ is in fact $\simeq(t-s)^2\langle|\mathbf{u}'|^2\rangle$ rather than $2(t-s)K$, in which case the integral diverges even if $N = 1$. It is concluded that the use of single particle statistics to model long-range scalar transport from an isolated source is justified in the sense that except at the source, the scalar variance is bounded even though $\kappa = 0$ and even if the source radius is vanishingly small.

The above calculation was simplified by the adoption of a normal profile for the source S as in (147), but the results are not dependent upon the choice. The difficulty at $\mathbf{x} = 0$ was noticed by *Durbin* [1980], but the satisfactory behavior elsewhere was not described. *Chatwin and Sullivan* [1979] demonstrated the existence of "core structures" in clouds of passive containments using general scaling arguments and also remarked on the adequacy of point source models in analyses of dispersion.

7. EFFECTIVE TOTAL DIFFUSIVITY FOR THE MEAN FIELD

In previous sections, second-order statistics of C have been examined: the variance spectrum $F(k, t)$ and the total variance

$\langle C^2 \rangle$. In this section the mean concentration $\langle C \rangle$ is considered. In particular, the asymptotic response for large time due to a nonrandom source $S(\mathbf{x}, t)$ will be estimated, and the results will be interpreted in terms of an effective total diffusivity.

Evolution equations for the mean concentration $\langle C \rangle$ have been derived by *Phythian and Curtis* [1978] and by *Drummond* [1982] using renormalized series expansions and Feynman path integral representations of (1), respectively. Both analyses led to equations of diffusion type. The effective total diffusivities were in both analyses less than the sum of the Lagrangian turbulent diffusivity $K = N^{-1}\mathrm{trace}(D_{ij})$, where D_{ij} is the integral in (34), plus the molecular diffusivity κ. This destructive interference between turbulent and molecular diffusion is evident in series solutions of (1) and (2) in powers of s derived by *Saffman* [1960] and *Okubo* [1967].

An estimate of $\langle C \rangle$ may be obtained directly, using the representation (5):

$$\langle C(\mathbf{x}, t)\rangle = \int_0^t ds \int d\mathbf{y}\ \langle G(\mathbf{x}-\mathbf{y}, t, s)S[\mathbf{A}(\mathbf{y}, t \mid s), s]\rangle \qquad (152)$$

where the average is over the turbulence only, since S is nonrandom. Let us assume that the infinitesimal line stretching in G and the single particle displacement $\mathbf{A}$ in S are statistically independent, since the former is due to integrated shear, while the latter is due to integrated velocity; this yields

$$\langle C(\mathbf{x}, t)\rangle = \int_0^t ds \int d\mathbf{y}\ \langle G(\mathbf{x}-\mathbf{y}, t, s)\rangle \int d\mathbf{z}\ S(\mathbf{z}, s)P(\mathbf{z}, s \mid \mathbf{y}, t) \qquad (153)$$

where $P(\mathbf{z}, s \mid \mathbf{y}, t)$ is the single particle displacement pdf defined by (27). For homogeneous turbulence, $P = P(\mathbf{z}-\mathbf{y}, s \mid t)$, and so (153) is a convolution product which has a simple representation in wave number space:

$$\langle \bar{C}(\mathbf{k}, t)\rangle = \int_0^t ds\ \langle \bar{G}(\mathbf{k}, t, s)\rangle \bar{S}(\mathbf{k}, s)\bar{P}(\mathbf{k}, s \mid t) \qquad (154)$$

where overbars denote Fourier transforms. (Previously, g was used in place of $\bar{G}$, but the latter symbol is introduced here for consistency.) Suppose further that $P(\mathbf{z}-\mathbf{y}, s \mid t)$ is normal, with variance $2(t-s)K$, as is asymptotically the case for isotropic turbulence for $t \to \infty$. Then $\bar{P} = e^{-Kk^2(t-s)}$. If molecular diffusion is neglected, then $\bar{G} \equiv 1$, and

$$\langle \bar{C}(\mathbf{k}, \infty)\rangle = (Kk^2)^{-1}\bar{S}(\mathbf{k}) \qquad (155)$$

for a steady source $S(\mathbf{x})$. If molecular diffusion is retained, but stretching is ignored, then $\bar{G} = e^{-\kappa k^2(t-s)}$, and (155) holds with K replaced by the effective total diffusivity $K_e = K + \kappa$. That is, there is no interference between molecular and turbulent diffusion.

Let us now retain stretching, using the simple model of *Batchelor* [1959] in which there is a nonrandom uniform shear field with a strain rate Ω (see section 5.2). Then the stretching factors grow exponentially in time, and

$$\bar{G}(\mathbf{k}, t, s) = \exp\left[-\frac{\kappa k^2}{2\Omega}\left(e^{2\Omega(t-s)} - 1\right)\right] \qquad (156)$$

Substitution of (156) into (154) with a steady source yields (155) with an effective total diffusivity

$$K_e = K + \kappa\left(1 + \frac{1}{K-1}\right) + O(\kappa^2 K^{-1}) \qquad (157)$$

which implies constructive interference between turbulent and molecular diffusion. Recall the assumption of independence of stretching and displacement and the use of the nonrandom uniform strain model.

Finally, assume that stretching is governed by a white noise strain rate, as in section 5.2. Then the results of *Bennett* [1986] may be used to show that

$$\langle \bar{G}(\mathbf{k}, t, s)\rangle \sim b[v(t-s)]^{-3/2} \exp\left[-\langle\alpha\rangle^2(t-s)/(4v)\right] \cdot (v^{1/2}\kappa^{1/2}k)^z K_0(v^{1/2}\kappa^{1/2}k) \qquad (158)$$

where $z = 1 - \frac{1}{2}\langle\alpha\rangle v^{-1}$ and K_0 is a modified Bessel function. For large x, $K_0(x) \sim bx^{-1/2}e^{-x}$. Note that the time and wave number dependence of $\bar{G}$ have separated asymptotically. Evidently, there is no longer an effective total diffusivity, according to the white noise strain model, and $\langle C(\mathbf{x}, \infty)\rangle$ does not satisfy anything like a steady state diffusion equation. It is concluded that calculations of corrections to the turbulent diffusivity K, which are after all only $O(\kappa/K) = O(Pe^{-1})$, are highly sensitive to model details. It should also be noted that the expansions used by *Phythian and Curtis* [1978] and also *Drummond* [1982] ensured that $\langle C \rangle$ satisfied a diffusion equation.

8. SUMMARY

An approximate analytical solution to the advection-diffusion equation has been used to estimate scalar variance spectra, total scalar variances, and mean scalar fields. The analysis is fundamentally Lagrangian in character.

The "-1" inertia-convective subrange, in isotropic stationary turbulence with an enstrophy-cascading inertial subrange, is deduced using *Lundgren*'s [1981] solution of the Richardson-Kraichnan equation with a separation-dependent relative diffusivity: the "2" law. The "$-\frac{5}{3}$" inertia-convective subrange, for an energy-cascading inertial subrange, is deduced from a new solution of the Richardson-Kraichnan equation with another separation-dependent relative diffusivity: the "$\frac{4}{3}$" law. The success of this calculation is shown to support the $\frac{4}{3}$ law over separation-independent alternatives. The -1 viscous-convective subrange, which exists for large Prandtl numbers, is derived by analogy with the (enstrophy) inertia-convective subrange. These spectral forms and relative diffusivities are found in oceanic and atmospheric data. An examination of the spectral representation of the relative diffusivity indicates that the transition from the $-\frac{5}{3}$ law to the -1 law should occur at wave numbers a decade smaller than the viscous cutoff k_V. This spectral misalignment is regularly observed. For low Prandtl number, nonuniversal inertia-diffusive subranges are found for enstrophy- and energy-cascading turbulence and isotropic scalar sources. However, the "-7" and "$-\frac{17}{3}$" forms are derived if the isotropic scalar sources are replaced with mean scalar gradients. The latter form has been observed in the laboratory. Joint statistics of separation and stretching are needed to describe viscous-diffusive subranges. These statistics are modeled using uniform strain fields which are deterministic, or have white noise time dependence, or have very long decorrelation times. There is a tendency toward essentially Gaussian spectral shapes at high wave numbers. However, the shapes are not universal, in agreement with recent oceanic observations.

Total scalar variances are estimated for several source configurations. The first is the isotropic source used in the analysis of spectral subranges. In the enstrophy inertia-convective

subrange there are equal contributions to the total variance from each decade; thus the total has a logarithmic dependence on the ill-defined upper and lower bounds in wave number space. The dynamics of the adjacent viscous-convective subrange are not well defined. In energy-cascading turbulence the total scalar variance is dominated by the contribution from the injection range. The total scalar variance in the presence of a mean scalar gradient, but no source, is found to have a finite value approximately independent of the molecular diffusivity for the scalar. Nevertheless, the analysis is crucially dependent upon the existence of a nonzero diffusivity, which causes the variance to depend on relative turbulent diffusion. The result just quoted holds for energy-cascading turbulence; for enstrophy-cascading turbulence the total variance has a mild dependence upon the molecular diffusivity, via the logarithm of the Peclet number. The total variance due to emission from a localized source is shown to be bounded for large time and independent of the source radius and molecular diffusivity, except at the source.

Finally, the total effective diffusivity for the mean scalar field is considered. The interaction between turbulent and molecular fields can, according to models presented here, be independently additive, or else constructive, or else such as to vitiate the concept of a total effective diffusivity altogether. Nevertheless, for large *Pe* the concept of a turbulent diffusivity must be essentially correct. That is, molecular diffusivity leads to an evolution equation for the mean scalar field which is not of diffusion type but must in some sense be close to that type.

The results surveyed here hold for isotropic turbulence. However, it should be noted that the approximate solution of the advection-diffusion equation and the pair displacement equation (equation (42)) (or its varients) are equally valid for inhomogeneous turbulence. Specialization was only necessary in sections 4–6, in order to make explicit estimates for relative and absolute (single particle) diffusivities. The problem of estimating the absolute diffusivity tensor for homogeneous but anisotropic turbulence was tackled directly by *Holloway and Kristmannsson* [1985]. They devised a second-order turbulence closure simultaneously for the β plane momentum equations and scalar diffusion equation. The closure was effected for the Eulerian forms of the equations; consequently, the scalar diffusivity tensor was expressed in terms of Eulerian velocity statistics rather than Lagrangian statistics. It was argued that the diffusivity tensor must be diagonal in east-west and north-south coordinates, while an approximate analysis showed that the north-south diffusivity decreased significantly with increasing values for the β parameter. It is no great oversimplification of this result to say that random Rossby waves are less effective in meridional diffusion than isotropic turbulence. The reduced meridional diffusivity is an expression of a decreased correlation between meridional velocity and the scalar field. A Lagrangian closure may lead to a different decorrelation.

Estimation of diffusivities for inhomogeneous turbulence is really beyond the scope of analytical theory. A recent one-dimensional analysis [*van Dop et al.*, 1985] considers absolute dispersion. As remarked in section 3, relative dispersion of particle pairs last only briefly in strongly inhomogeneous turbulence.

APPENDIX I: PARTICLE DISPERSION IN A SHEAR FLOW

Suppose there is a mean shear in the velocity fields, $\langle \mathbf{u} \rangle = (\gamma x_3, 0, 0)$, and suppose that only a transverse velocity component fluctuates, $\langle u_1'^2 \rangle = \langle u_2'^2 \rangle = 0$ and $\langle u_3'^2 \rangle \neq 0$. Consider two particles with an initial separation $\mathbf{R} = (r_1, 0, 0)$ sufficiently large in magnitude so that their motions are independent. Their motions may be regarded as independent random walks, so the evolution of the components of their separation may be modeled by

$$dR_1 = \gamma R_3 \, dt \tag{A1}$$

$$dR_2 = 0 \tag{A2}$$

$$dR_3 = 2K^{1/2} \, d\omega\,(t) \tag{A3}$$

where $\omega(t)$ is the Wiener process, while K is a constant diffusivity (for stationary homogeneous turbulence). It follows easily that $\langle R_3^2 \rangle = 4Kt$, and $\langle R_1 R_3 \rangle = 2K\gamma t^2$, so

$$\langle R_1^2 \rangle = r_1^2 + \tfrac{4}{3} K\gamma^2 t^3 \tag{A4}$$

This t^3 law for particle separation is widely observed [*Okubo*, 1971], but (A4) is not a consequence of energy-cascading inertial range scaling.

APPENDIX II: INFINITESIMAL STRETCHING

By definition of $\mathbf{X} = \mathbf{A}(\mathbf{x}, t \mid s)$, there is the identity

$$\mathbf{x} = \mathbf{A}[\mathbf{A}(\mathbf{x}, t \mid s), s \mid t] \tag{A5}$$

Hence

$$\delta_{ij} = \frac{\partial A_j}{\partial X_l}(\mathbf{X}, s \mid t) \frac{\partial A_l}{\partial x_i}(\mathbf{x}, t \mid s) \tag{A6}$$

which yields

$$k_i = h_l \frac{\partial A_l}{\partial x_i}(\mathbf{x}, t \mid s) \tag{A7}$$

where

$$h_l = k_j \frac{\partial A_j}{\partial X_l}(\mathbf{X}, s \mid t) \tag{A8}$$

is the stretching vector introduced in section 5.2. From (A7) and (3) it follows that

$$\frac{\partial h_l}{\partial s} \frac{\partial A_l}{\partial x_i}(\mathbf{x}, t \mid s) + h_l\left(\frac{\partial}{\partial x_i}\right) u_l(\mathbf{x}, t \mid s) = 0 \tag{A9}$$

which becomes

$$\frac{\partial h_l}{\partial s} \frac{\partial A_l}{\partial x_i}(\mathbf{x}, t \mid s) + h_l\left(\frac{\partial u_l}{\partial x_m}\right)(\mathbf{x}, t \mid s) \frac{\partial A_m}{\partial x_i}(\mathbf{x}, t \mid s) = 0 \tag{A10}$$

Analogous to (A6), there is the identity

$$\delta_{ij} = \frac{\partial A_j}{\partial x_p}(\mathbf{x}, t \mid s) \frac{\partial A_p}{\partial X_i}(\mathbf{X}, s \mid t) \tag{A11}$$

so multiplying (A10) by $(\partial A_i / \partial X_n)$ yields

$$\frac{\partial h_n}{\partial s} = -h_l \frac{\partial u_l}{\partial x_n} \tag{A12}$$

or

$$\frac{\partial}{\partial s} \mathbf{h} = -\mathbf{W}^T \mathbf{h} \tag{113}$$

where

$$W_{ij} = \frac{\partial u_i}{\partial x_j}(\mathbf{x}, t \mid s) \tag{114}$$

Section 5

Γ	mean scalar gradient (see equation (107)).
$V_{uu}(\mathbf{r})$	Eulerian velocity covariance at separation $\mathbf{r}$. (see after equation (108)).
$k_D \equiv k_V\, Pr^{3/4}$	lower limit of inertia-diffusive subrange ($Pr \ll 1$).
W_{ij}	velocity shear tensor (see equation (114)).
α	random strain rate, with mean $\langle\alpha\rangle$ and variance v^2.
$q = \Omega\langle\alpha\rangle^{-1}$	constant in universal form of *Batchelor* [1959] spectrum (see equation (125) and after equation (130)).

Section 6

$U = (\varepsilon m^{-1})^{1/3}$	root-mean square turbulent velocity.
$Pe \equiv U\kappa^{-1}m^{-1}$	Peclet number.
$Re \equiv U\nu^{-1}m^{-1}$	Reynolds number.
V_{uu}	Lagrangian velocity covariance (see equation (139)).

Note that other symbols not listed have also been used only locally or as dummy variables; b and b' always signify dimensionless constants.

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The expression (equation (18)) for the variance spectrum may be written as

$$F(k, t) = \chi \int da\,(\mathbf{k})a(l)^{-1} \int da\,(\mathbf{l}) \int_0^t ds \int d\mathbf{r}\, \langle e^{i(\mathbf{k}\cdot\mathbf{r}-\mathbf{l}\cdot\mathbf{R})} g^{2} \rangle \tag{A13}$$

where $\mathbf{R}$ and g are random variables satisfying (116) and (117), respectively. It follows from (116) that $\mathbf{R}$ is given by

$$\mathbf{R} = \mathsf{E}(t \mid s)\mathbf{r} \tag{A14}$$

where

$$\frac{\partial \mathsf{E}}{\partial s} = \mathsf{W}\mathsf{E} \tag{A15}$$

and $\mathsf{E}(t \mid t) = \mathsf{I}$, the identity matrix. Substituting (A14) into (A13) and integrating over $\mathbf{r}$ yields

$$F(k, t) = \chi \int da\,(\mathbf{k})a(l)^{-1} \int da\,(\mathbf{l}) \int_0^t ds\, \langle \delta[\mathbf{k} - \mathsf{E}^T(t \mid s)\mathbf{l}] g^2 \rangle \tag{A16}$$

Now g is given by

$$g = \exp\left[-\kappa \int_s^t h^2(t \mid s')\, ds' \right] \tag{A17}$$

where by (113),

$$\mathbf{h}(t \mid s') = \mathsf{E}^T(s' \mid t)\mathbf{k} \tag{A18}$$

The expectation in (A16) is the variance of g, conditional upon

$$\mathbf{k} = \mathsf{E}^T(t \mid s)\mathbf{l} \tag{A19}$$

Using (A17)–(A19) and the identity

$$\mathsf{E}(s' \mid s) = \mathsf{E}(s' \mid t)\mathsf{E}(t \mid s) \tag{A20}$$

shows that the expectation in (A16) may be written as $\langle g^2 \rangle$ where

$$g_t = -\kappa k^2 g \qquad \mathbf{k}_t = \mathsf{W}^T\mathbf{k} \tag{119}$$

subject to $g = 1$ and $\mathbf{k} = \mathbf{l}$ at $s = t$.

NOTATION

This is a list of frequently occurring symbols, in order of introduction.

Section 2

$\mathbf{x}, \mathbf{X}$	N-dimensional space coordinates.
t, s	time variables.
$\mathbf{u} = \mathbf{u}(\mathbf{x}, t)$	N-dimensional fluid velocity field.
κ	scalar molecular diffusivity.
$C = C(\mathbf{x}, t)$	scalar concentration field.
$S = S(\mathbf{x}, t)$	scalar source field.
$\mathbf{A} = \mathbf{A}(\mathbf{x}, t \mid s)$	position at time s of a particle known to pass through position $\mathbf{x}$ at time t (see equation (3)).
$G(\mathbf{x} - \mathbf{y}, t, s)$	fundamental solution of advection-diffusion equation in Lagrangian form (equation 4)), assuming spatially uniform transformation factors.
$\mathbf{k}, \mathbf{l}, \mathbf{m}$	N-dimensional wave number vectors. Scalar variance is injected at $l = \lvert\mathbf{l}\rvert$; kinetic energy is injected at $m = \lvert\mathbf{m}\rvert$.
$g(\mathbf{k}, t, s)$	Fourier transform of $G(\mathbf{x}, t, s)$ (see equation (6)).
Q, L	transformation or stretching factors (see equation (6)).
$\mathbf{D}, \mathbf{r}, \mathbf{R}$	N-dimensional separation vectors.
$\langle\ \rangle, '$	ensemble average and fluctuation.
$V = \langle C(\mathbf{x}, t)^2 \rangle_{u,S}$	total scalar variance, averaged over turbulence and sources.
$da\,(\mathbf{k})$	in $\mathbf{k}$ space; area element on an N sphere of radius $k = \lvert\mathbf{k}\rvert$.
$F(k, t)$	variance spectrum of scalar concentration (see equation (10)).
$a(k)$	area of N sphere of radius k (see equation (11)).
$\mathscr{B}(kD)$	spherical average of $e^{i\mathbf{k}\cdot\mathbf{D}}$ (see equation (12)).
$P(\cdots, \cdots, s \mid \cdots, \cdots, t)$	pdf of values at time s, conditional on their taking values at time t.
χ	intensity of scalar source S (see equations (15) and (16).

Section 3

$p(\cdots, \cdots, s \mid \cdots, \cdots, t)$	"micro" distribution function for a given realization: $\langle p \rangle = P$ (see equations (25) and (27)).
$D_{ij}(\mathbf{x}, \mathbf{y}, t \mid s)$	Lagrangian diffusivity tensor (equation (43)).
η_{ij}	Lagrangian relative diffusivity tensor (see equation (49)).
η	longitudinal component of η_{ij}.
ζ	Richardson's relative diffusivity (see equation (55)).
$K_{ij}(\mathbf{X}, \mathbf{X}, s \mid t)$	Taylor's diffusivity tensor (see equation (67)).
$\omega(t)$	N-dimensional Wiener process, with uncorrelated components and uncorrelated increments.

Section 4

$E(k)$	kinetic energy spectrum (see equation (69)).
ε	mean dissipation rate of turbulent kinetic energy.
ν	kinematic viscosity.
$k_V \equiv \varepsilon^{1/4}\nu^{-3/4}$	Kolmogorov wave number.
K_V	Kolmogorov constant (see equation (70)).
$\mathscr{F} = \mathscr{F}(\theta)$	high-pass filter due to geometry of isotropic turbulence (see equations (74) and (75)).
$\mathscr{L} = \mathscr{L}(k, w)$	dimensionless Lagrangian spectrum of kinetic energy (see equations (76) and (77)).
λ	mean dissipation rate of turbulent vertical vorticity variance (enstrophy).
$V(r)$	equilibrium scalar covariance at separation $r = \lvert\mathbf{r}\rvert$.
$F(k)$	equilibrium scalar variance spectrum.
$\Omega \equiv (\varepsilon/\nu)^{1/2}$	root-mean-square velocity shear.
$Pr = \nu/\kappa$	Prandtl number.
$k_B \equiv k_V\, Pr^{1/2} = (\Omega\kappa^{-1})^{1/2}$	*Batchelor*'s [1959] cutoff wave number.

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